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ALMQVIST & WIKSELL / STOCKHOLM

1957

The crystal structure of finnemanite $\text{Pb}_5\text{Cl}(\text{AsO}_3)_3$

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With 1 figure in the text

Previous investigations

The rare mineral finnemanite, as yet only found in Långban, Sweden, has been described by G. AMINOFF (1923). Later AMINOFF and A. L. PARSONS (1927) found by studying the Laue symmetry, that finnemanite belonged to either $6/m$ or 6 . The symmetry class $\bar{6}$ also gave a Laue pattern of the same symmetry but was excluded owing to the marked hexagonal development of the finnemanite crystals.

The lattice dimensions of the unit cell of finnemanite were: $a = 10.21 \text{ \AA}$ and $c = 6.97 \text{ \AA}$. AMINOFF and PARSONS found the cell dimensions of finnemanite to be very close to those of mimetite $\text{Pb}_5\text{Cl}(\text{AsO}_4)_3$, which subsequently had been assumed to be structurally related to finnemanite. However, although the lattice dimensions are remarkably close to each other, AMINOFF and PARSONS found marked differences in the intensities of the photograms of the two minerals and they assumed therefore, that the differences might be due to different atomic arrangements in the two minerals. Especially they stressed the importance of the presence of trivalent arsenic in finnemanite as against pentavalent arsenic in mimetite.

Material for the present investigations

A small prismatic crystal, elongated parallel to the c -axis, was used for the present investigation. The crystal was cut and ground approximately into a cylinder around the c -axis.

Unit cell, space group and atomic parameters of the heavy atoms

Equi-inclination Weissenberg photographs were taken around the c -axis using unfiltered iron radiation. The Weissenberg photographs obtained registered the reflections $(h k i 0)$, $(h k i 1)$ and $(h k i 2)$. The lattice constants calculated are: $a = 10.28 \text{ \AA}$ and $c = 7.00 \text{ \AA}$, which values are close to the data found by AMINOFF. The unit cell contains two molecules $\text{Pb}_5\text{Cl}(\text{AsO}_3)_3$. Thus 10 lead, 2 chlorine, 6 arsenic and 18 oxygen atoms are to be placed into the cell.

According to AMINOFF and PARSONS finnemanite belongs to either $6/m$ or 6 . In $6/m$ there are two space groups:

C_{6h}^1 -P $6/m$ with horizontal symmetry planes in $z = 0$ and $\frac{1}{2}$.

C_{6h}^2 -P $6_3/m$ with horizontal symmetry planes in $z = \frac{1}{4}$ and $\frac{3}{4}$ and sixfold screw axis in $x = y = 0$.

Table 1. (According to Int. Tab. 1952.)

Atoms	Possible positions	
	$C_{6h}^1-P 6/m$	$C_{6h}^2-P 6_3/m$
$\left. \begin{array}{l} 2 Pb_I \\ 2 Pb_{II} \\ 2 Cl \end{array} \right\}$	(a) $0,0,0 + (b) 0,0,\frac{1}{2}$ (c) $\frac{1}{3},\frac{2}{3},0; \frac{2}{3},\frac{1}{3},0$ (d) $\frac{1}{3},\frac{2}{3},\frac{1}{2}; \frac{2}{3},\frac{1}{3},\frac{1}{2}$	(a) $0,0,\frac{1}{4}; 0,0,\frac{3}{4}$ (c) $\frac{1}{3},\frac{2}{3},\frac{1}{4}; \frac{2}{3},\frac{1}{3},\frac{3}{4}$ (d) $\frac{1}{3},\frac{2}{3},\frac{3}{4}; \frac{2}{3},\frac{1}{3},\frac{1}{4}$
$\left. \begin{array}{l} 6 Pb_{III} \\ 6 As \end{array} \right\}$	(i) $\frac{1}{2},0,z; 0,\frac{1}{2},z; \frac{1}{2},\frac{1}{2},z; \frac{1}{2},0,\bar{z}; 0,\frac{1}{2},\bar{z};$ $\frac{1}{2},\frac{1}{2},\bar{z}$ or (j) $x,y,0; \bar{y},x-y,0; y-x,\bar{x},0; \bar{x},\bar{y},0;$ $y,y-x,0; x-y,x,0$ or (k) $x,y,\frac{1}{2}; \bar{y},x-y,\frac{1}{2}; y-x,\bar{x},\frac{1}{2}; \bar{x},\bar{y},\frac{1}{2};$ $y,y-x,\frac{1}{2}; x-y,x,\frac{1}{2}$	(g) $\frac{1}{2},0,0; 0,\frac{1}{2},0; \frac{1}{2},\frac{1}{2},0; \frac{1}{2},0,\frac{1}{2}; 0,\frac{1}{2},\frac{1}{2};$ $\frac{1}{2},\frac{1}{2},\frac{1}{2}$ or (h) $x,y,\frac{1}{4}; \bar{y},x-y,\frac{1}{4}; y-x,\bar{x},\frac{1}{4}; \bar{x},\bar{y},\frac{3}{4};$ $y,y-x,\frac{3}{4}; x-y,x,\frac{3}{4}$
Atoms	Possible positions	
	$C_6^1-P 6$	$C_6^6-P 6_3$
$\left. \begin{array}{l} 2 Pb_I \\ 2 Pb_{II} \\ 2 Cl \end{array} \right\}$	2 (a) $0,0,z$ (b) $\frac{1}{3},\frac{2}{3},z; \frac{2}{3},\frac{1}{3},z$ (b) $\frac{1}{3},\frac{2}{3},z; \frac{2}{3},\frac{1}{3},z$	(a) $0,0,z; 0,0,\frac{1}{2}+z$ (b) $\frac{1}{3},\frac{2}{3},z; \frac{2}{3},\frac{1}{3},\frac{1}{2}+z$ (b) $\frac{1}{3},\frac{2}{3},z; \frac{2}{3},\frac{1}{3},\frac{1}{2}+z$
$\left. \begin{array}{l} 6 Pb_{III} \\ 6 As \end{array} \right\}$	(d) $x,y,z; \bar{y},x-y,z; y-x,\bar{x},z; \bar{x},\bar{y},z;$ $y,y-x,z; x-y,x,z$	(c) $x,y,z; \bar{y},x-y,z; y-x,\bar{x},z; \bar{x},\bar{y},\frac{1}{2}+z;$ $y,y-x,\frac{1}{2}+z; x-y,x,\frac{1}{2}+z$

Table 2. The final parameters of the atomic positions in Finnemanite.
Space group $C_6^6-P 6_3$.

2 Pb _I in (a)	$x=0$	$y=0$	$z=0$
2 Pb _{II} „ (b)	$x=\frac{1}{3}$	$y=\frac{2}{3}$	$z=0.620$
2 Cl „ (b)	$x=\frac{1}{3}$	$y=\frac{2}{3}$	$z=0.120$
6 Pb _{III} „ (c)	$x=0.300$	$y=0.400$	$z=0.250$
6 As „ (c)	$x=0.270$	$y=0.370$	$z=0.770$
6 O _I „ (c)	$x=0.205$	$y=0.170$	$z=0.750$
6 O _{II} „ (c)	$x=0.145$	$y=0.440$	$z=0.820$
6 O _{III} „ (c)	$x=0.375$	$y=0.445$	$z=0.550$

Both these space groups have two- and sixfold positions and are therefore possible. The atomic arrangement in these space groups is shown in Table 1.

In the crystal class 6 there are six space groups. The space groups $P 6_1$, $P 6_2$, $P 6_4$ and $P 6_5$ have only three- and sixfold positions. They must therefore be excluded, because the number of lead and chlorine atoms do not fit the atomic positions, and a statistical distribution is unlikely. The remaining two space groups, $P 6$ and $P 6_3$, having two- and sixfold positions, are possible. The atomic positions of $P 6$ and $P 6_3$ are shown in Table 1.

Table 3. Weissenberg photograph of Finnemanite. Rotation axis [0001]. Zero layer line. Fe-radiation. Comparison between observed and calculated intensities.

$h k i l$	$\frac{\sin \theta}{\lambda}$	F obs.	F calc.	$h k i l$	$\frac{\sin \theta}{\lambda}$	F obs.	F calc.
0 3 $\bar{3}$ 0	0.171	70	75	3 2 $\bar{5}$ 0	0.247	50	36
0 4 $\bar{4}$ 0	0.227	30	24	3 3 $\bar{6}$ 0	0.294	< 20	11
0 5 $\bar{5}$ 0	0.285	20	17	3 4 $\bar{7}$ 0	0.346	20	9
0 6 $\bar{6}$ 0	0.341	< 10	7	3 5 $\bar{8}$ 0	0.398	15	19
0 7 $\bar{7}$ 0	0.398	< 5	17	3 6 $\bar{9}$ 0	0.450	< 5	10
0 8 $\bar{8}$ 0	0.455	< 5	0	3 7 $\bar{10}$ 0	0.504	5	24
1 2 $\bar{3}$ 0	0.151	25	22	4 1 $\bar{5}$ 0	0.261	40	33
1 3 $\bar{4}$ 0	0.205	15	23	4 2 $\bar{6}$ 0	0.299	40	32
1 4 $\bar{5}$ 0	0.261	100	87	4 3 $\bar{7}$ 0	0.346	< 10	13
1 5 $\bar{6}$ 0	0.316	20	25	4 4 $\bar{8}$ 0	0.393	25	64
1 6 $\bar{7}$ 0	0.372	15	6	5 1 $\bar{6}$ 0	0.316	30	50
1 7 $\bar{8}$ 0	0.430	35	78	5 2 $\bar{7}$ 0	0.355	35	55
1 8 $\bar{9}$ 0	0.486	10	1	5 3 $\bar{8}$ 0	0.398	25	19
2 1 $\bar{3}$ 0	0.151	45	90	5 4 $\bar{9}$ 0	0.444	< 5	8
2 2 $\bar{4}$ 0	0.196	35	29	5 5 $\bar{10}$ 0	0.492	10	8
2 3 $\bar{5}$ 0	0.247	20	18	6 1 $\bar{7}$ 0	0.372	15	16
2 4 $\bar{6}$ 0	0.299	30	56	6 2 $\bar{8}$ 0	0.410	< 15	16
2 5 $\bar{7}$ 0	0.355	15	13	6 3 $\bar{9}$ 0	0.450	15	37
2 6 $\bar{8}$ 0	0.410	< 5	14	7 1 $\bar{8}$ 0	0.429	5	8
2 7 $\bar{9}$ 0	0.465	15	9	7 2 $\bar{9}$ 0	0.465	10	37
3 1 $\bar{4}$ 0	0.205	20	35				

Table 4. Weissenberg photograph of Finnemanite. Rotation axis [0001]. First layer line. Fe-radiation. Comparison between observed and calculated intensities.

$h k i l$	$\frac{\sin \theta}{\lambda}$	F obs.	F calc.	$h k i l$	$\frac{\sin \theta}{\lambda}$	F obs.	F calc.
0 3 $\bar{3}$ 1	0.184	< 20	8	3 1 $\bar{4}$ 1	0.216	25	14
0 4 $\bar{4}$ 1	0.237	< 25	37	3 2 $\bar{5}$ 1	0.257	60	43
0 5 $\bar{5}$ 1	0.292	20	13	3 3 $\bar{6}$ 1	0.302	35	27
0 6 $\bar{6}$ 1	0.347	30	37	3 4 $\bar{7}$ 1	0.351	10	20
0 7 $\bar{7}$ 1	0.402	10	11	3 5 $\bar{8}$ 1	0.422	5	17
0 8 $\bar{8}$ 1	0.458	< 5	13	3 6 $\bar{9}$ 1	0.472	15	27
1 2 $\bar{3}$ 1	0.166	35	36	4 1 $\bar{5}$ 1	0.269	15	10
1 3 $\bar{4}$ 1	0.216	< 25	12	4 2 $\bar{6}$ 1	0.308	< 15	16
1 4 $\bar{5}$ 1	0.269	< 20	5	4 3 $\bar{7}$ 1	0.351	< 10	9
1 5 $\bar{6}$ 1	0.323	< 15	22	4 4 $\bar{8}$ 1	0.398	10	6
1 6 $\bar{7}$ 1	0.398	< 5	13	5 1 $\bar{6}$ 1	0.323	15	17
1 7 $\bar{8}$ 1	0.450	< 5	0	5 2 $\bar{7}$ 1	0.360	< 10	9
2 1 $\bar{3}$ 1	0.166	50	20	5 3 $\bar{8}$ 1	0.402	< 5	22
2 2 $\bar{4}$ 1	0.209	< 25	4	6 1 $\bar{7}$ 1	0.378	15	40
2 3 $\bar{5}$ 1	0.257	20	10	6 2 $\bar{8}$ 1	0.414	< 5	13
2 4 $\bar{6}$ 1	0.308	30	13	6 3 $\bar{9}$ 1	0.455	< 5	9
2 5 $\bar{7}$ 1	0.360	30	35	7 1 $\bar{8}$ 1	0.450	5	8
2 6 $\bar{8}$ 1	0.414	< 5	20	7 2 $\bar{9}$ 1	0.485	15	8

Table 5. Weissenberg photograph of Finnemanite. Rotation axis $[0001]$. Second layer line. Fe-radiation. Comparison between observed and calculated intensities.

$h k i l$	$\frac{\sin \theta}{\lambda}$	$ F $ obs.	$ F $ calc.	$h k i l$	$\frac{\sin \theta}{\lambda}$	$ F $ obs.	$ F $ calc.
0 3 $\bar{3}$ 2	0.222	20	22	3 1 $\bar{4}$ 2	0.250	< 20	14
0 4 $\bar{4}$ 2	0.268	50	50	3 2 $\bar{5}$ 2	0.285	< 20	53
0 5 $\bar{5}$ 2	0.317	40	42	3 3 $\bar{6}$ 2	0.327	45	47
0 6 $\bar{6}$ 2	0.368	35	51	3 4 $\bar{7}$ 2	0.372	< 10	39
0 7 $\bar{7}$ 2	0.421	10	9	3 5 $\bar{8}$ 2	0.430	< 5	10
0 8 $\bar{8}$ 2	0.473	10	13	3 6 $\bar{9}$ 2	0.471	15	32
1 3 $\bar{4}$ 2	0.250	50	54	4 1 $\bar{5}$ 2	0.296	45	39
1 4 $\bar{5}$ 2	0.296	< 20	30	4 2 $\bar{6}$ 2	0.332	< 15	20
1 5 $\bar{6}$ 2	0.346	< 15	18	4 3 $\bar{7}$ 2	0.372	30	36
1 6 $\bar{7}$ 2	0.397	< 10	16	4 4 $\bar{8}$ 2	0.417	15	20
2 2 $\bar{4}$ 2	0.267	40	38	4 5 $\bar{9}$ 2	0.464	< 5	30
2 3 $\bar{5}$ 2	0.285	30	11	5 1 $\bar{6}$ 2	0.346	< 15	25
2 4 $\bar{6}$ 2	0.332	< 15	29	5 2 $\bar{7}$ 2	0.380	< 10	18
2 5 $\bar{7}$ 2	0.380	30	45	5 3 $\bar{8}$ 2	0.430	30	42
2 6 $\bar{8}$ 2	0.432	10	32	5 4 $\bar{9}$ 2	0.464	10	24

It is evident from Table 1 that the projections of the heavy atoms on (0001) in all four space groups are identical.

The x - and y -parameters of the heavy atoms lead and arsenic have been determined by means of the trial-and-error method by systematic variation of the parameters, accompanied by a comparison between calculated and observed intensities of $(hki0)$ reflections. Owing to the large absorption especially at low angles the observed intensities have been approximately corrected for the absorption according to a method, described by G. ALBRECHT (1939), assuming a perfect cylindrical form of the crystal. The final x - and y -parameters of these atoms are shown in Table 2, and a comparison between calculated and observed intensities, obtained with these parameters, is given in Table 3.

So far the one space group is indistinguishable from the other, but by finding the z -parameters it is possible to determine it uniquely. The z -parameters of lead and arsenic have also been determined by the trial-and-error method, using the $(hki1)$ and $(hki2)$ reflections. The observed intensities of the first and second layer lines have also been roughly corrected according to the method of G. ALBRECHT, mentioned above. A comparison between the calculated and observed intensities of $(hki1)$ and $(hki2)$, obtained with the final z -parameters of lead and arsenic, is given in Tables 4 and 5.

The values of the z -parameters of lead and arsenic are shown in Table 2. These z -parameters indicate atomic positions in two xy -planes around a screw axis in $x=y=0$ and parallel to the c -direction. In $P6/m$ and $P6$ there are no screw axes, and therefore these two space groups must be excluded. In $P6_3/m$ and $P6_3$ the sixfold positions of lead (Pb_{III}) and arsenic are possible, if they are arranged in two planes around the screw axis. The z -parameters of these atoms are found to give positions not equal to $z = \frac{1}{4}$ and $\frac{3}{4}$, which is a necessary condition if the space group were $P6_3/m$. Thus $P6_3$ is the only possible space group.

Table 6. Interatomic distances in finnemanite.

Atom	Coordination	Number of atoms	Kinds of atoms	Distance (Å)
Pb _I	6 octahedron	6	O _I	2.63
Pb _{II}	8	3	O _{II}	2.56
		3	O _{III}	2.58
		2	Cl	3.50
Pb _{III}	4 tetrahedron	1	O _I	2.45
		1	O _{II}	2.24
		1	O _{III}	2.21
		1	Cl	2.75
As	3 pyramid	1	O _I	1.83
		1	O _{II}	1.81
		1	O _{III}	1.80

Interatomic distances between the oxygen atoms in pyramidal coordination with As.

O _I			O _{II}	3.14
O _I			O _{III}	2.84
O _{II}			O _{III}	3.01

Interatomic distances As-Pb.

As		1	Pb _{II}	2.97
		1	Pb _{III}	3.36
		1	Pb _{III}	3.64
		1	Pb _{III}	3.52

Atomic parameters of oxygen and chlorine

Owing to the insignificant diffraction power of the oxygen and chlorine atoms compared with especially the lead atoms the former have a very small influence on the reflection intensities. The positions of the chlorine and oxygen atoms have therefore been fixed on spatial considerations.

The two chlorine atoms are assumed to occupy the twofold position (b). Position (a) is impossible for space reasons because two lead atoms (Pb_I) occupy it already. The eighteen oxygen atoms occupy three sixfold positions (c).

The coordination of oxygen around arsenic is important for the calculation of the parameters of the oxygen atoms. The proportion one arsenic to three oxygen atoms might be obtained in two different ways:

1. Four oxygen atoms in tetrahedral configuration.
2. Three oxygen atoms in triangular (planar or pyramidal) configuration.

Oxygen atoms arranged in octahedra or trigonal prisms around arsenic are both impossible arrangements, because the octahedra or trigonal prisms cannot be placed in the unit cell with all corners shared, which is necessary in order to have the proportion AsO₃.

Alternative 1. Four-coordinated arsenic in tetrahedral coordination with oxygen

The tetrahedra have two corners shared, giving the proportion AsO_3 . The tetrahedra are found to form an infinite chain, which on account of the presence of a screw axis along the *c*-axis is arranged in a spiral around the lead atoms in position (a). Attempts have been made to group the oxygen atoms according to this system, but the distance As–O seems improbably large (2.39, 2.20 and 2.03 Å). Assuming mainly covalent bonds the distance As–O is about 1.84 Å, using PAULING and HUGGINS' values of tetrahedral covalent radii of arsenic and oxygen.

Alternative 2. Three-coordinated arsenic in pyramidal coordination with oxygen

It is possible to adapt the distances As–O to the length of covalent bonds, if a pyramidal arrangement of three oxygen atoms around arsenic is assumed. The mean estimated distance of As–O, is 1.82 Å, while the length of normal covalent bonds As–O is 1.84 Å according to PAULING and HUGGINS (1934). Considering the distances between oxygen and adjacent atoms of lead and chlorine, the most suitable parameters of the oxygen atoms are those, given in Table 2.

Description of the structure

In Fig. 1 the structure of finnermanite is shown in projection on (0 0 0 1).

The six arsenic atoms are arranged in two planes around the origin. Three oxygen atoms form a pyramidal configuration around the arsenic atom with the distances As–O = 1.80, 1.81 and 1.83 Å.

In all known structures of oxygen compounds with trivalent arsenic, the oxygen configuration around the arsenic has been found to be pyramidal. According to R. M. BOZORTH (1923) cubic As_2O_3 has a molecular structure consisting of As_4O_6 molecules, which are built up of four pyramidal AsO_3 groups. The oxygen atoms of these groups are shared between two AsO_3 . BOZORTH found the distance As–O to be 2.01 Å, which later was refined by K. E. ALMIN and A. WESTGREN (1943) to 1.80 ± 0.05 Å.

W. H. ZACHARIASEN (1931) has suggested the pyramidal structure of AsO_3 (and further SbO_3 , SO_3 , ClO_3 and BrO_3) to be caused by the presence of two free valency electrons. These electrons occupy one corner of a tetrahedral configuration around arsenic, the three remaining corners being occupied by oxygen.

The pyramidal configuration of the oxygen around the arsenic is also found in an isomorphous series of antimonites and arsenites of the type AB_2O_4 , where A = Mg, Mn, Fe, Co, Ni, Zn and B = As, Sb. These compounds were investigated by S. STÅHL (1943). They were found to be built up in the same way as in minium, Pb_3O_4 , the structure of which was somewhat previously determined by A. BYSTRÖM and A. WESTGREN (1943) simultaneously with S. T. GROSS (1943). The AB_2O_4 compounds are composed of AO_6 octahedra in chains, held together laterally by flat BO_3 pyramids.

Later J. ZEMANN (1950, 1951) found that schafarzikite ($FeSb_2O_4$) and trippkeite ($CuAs_2O_4$) were isostructural with the antimonites and arsenites, investigated by S. STÅHL. The distance As–O in trippkeite was calculated to be 1.82 Å (mean distance). A. BYSTRÖM (1947) has suggested mainly covalent bonds in the AsO_3 groups of the compounds of the type AB_2O_4 . According to him, arsenic has an inert pair of valency electrons, which is stereochemically active. The coordination might hence be described as tetrahedral with oxygen atoms in three corners and the inert pair of elec-

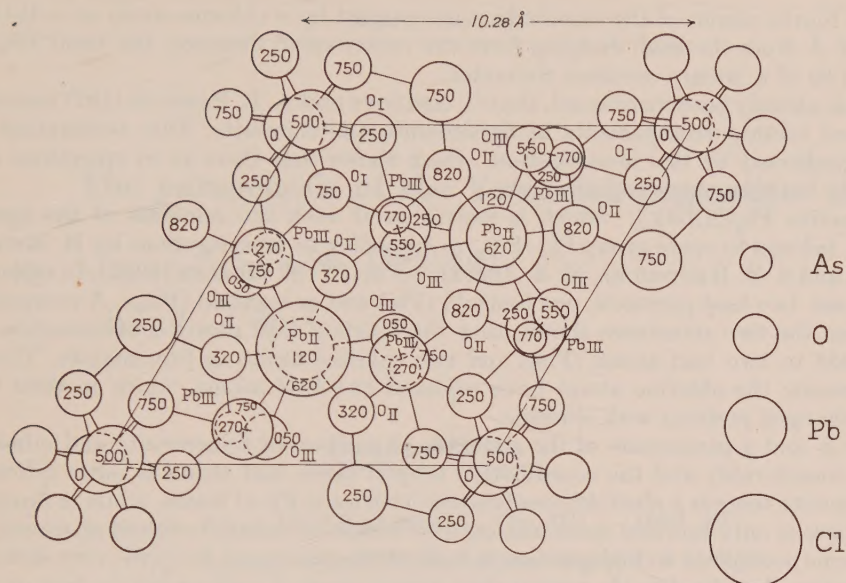


Fig. 1. The structure of finnemanite, projected on (0 0 0 1). Superimposed Pb-atoms in $x = y = 0$ are symmetrically displaced.

trons occupying the fourth corner in the same way as was assumed in As_2O_3 by ZACHARIASEN.

If we assume mainly covalent bonds between arsenic and oxygen in the AsO_3 groups of finnemanite (mean value $\text{As}-\text{O} = 1.82 \text{ \AA}$), there might be the same tetrahedral configuration around arsenic with three corners occupied by oxygen and the inert pair of electrons in arsenic directed towards the fourth corner.

From Fig. 1 it is evident that the lead atoms occupy three different positions (Pb_I , Pb_{II} and Pb_{III}). Two lead atoms (Pb_I) are surrounded by six oxygen atoms (O_I) in a regular octahedral configuration. The octahedra share three corners with the adjacent octahedra, forming in this way chains parallel to the c -axis. The oxygen atoms are fixed at a distance from lead, being the same as the interionic bond $\text{Pb}^{+2}-\text{O}^{-2} = 2.63 \text{ \AA}$, according to the value given by PAULING.

The lead atoms (Pb_{II}) in twofold position are surrounded by six oxygen atoms and two chlorine atoms. The distances $\text{Pb}_{II}-\text{O}$ (2.56 and 2.58 \AA) are somewhat shorter than the normal interionic bond $\text{Pb}-\text{O}$ with lead in 8-coordination (according to A. BYSTRÖM 2.73 \AA). The distance $\text{Pb}_{II}-\text{Cl}$ (3.50 \AA) is on the contrary considerably longer than the normal interionic bond (according to BYSTRÖM 3.02 \AA).

An entirely different coordination is found around lead in sixfold position (Pb_{III}). It is surrounded by three oxygen and one chlorine atom. The three oxygen atoms and the chlorine atom form a somewhat distorted tetrahedron. The distances $\text{Pb}_{III}-\text{O}$ are calculated to be 2.21, 2.24 and 2.45 \AA . The length of the purely covalent bond in tetrahedral configuration is $1.46 (\text{Pb}) + 0.66 (\text{O}) = 2.12 \text{ \AA}$, according to PAULING and HUGGINS. Using Wells' value (1949) of the covalent radius of oxygen, the distance $\text{Pb}-\text{O}$ would be $1.46 + 0.74 = 2.20 \text{ \AA}$. The mean distance of $\text{Pb}_{III}-\text{O}$ in finnemanite is 2.30 \AA , which indicates bonds of mainly covalent nature.

The fourth corner of the tetrahedron is occupied by a chlorine atom at a distance of 2.75 Å from the lead. Judging from the rather small distance, the bond $Pb_{III}-Cl$ should be of a mainly covalent character.

It has already been mentioned, that G. AMINOFF and A. L. PARSONS (1927) assumed different atomic arrangements in finnemanite and mimetite. This assumption has been confirmed by this investigation, which shows that there is no structural relationship between these two minerals.

Mimetite $Pb_5Cl(AsO_4)_3$, which is isostructural with the minerals of the apatite group, belongs to space group $C_{6h}^2-P 6_3/m$ according to investigations by H. MEHMEL (1931) and S. B. HENDRICKS, M. E. JEFFERSON and V. M. MOSLEY (1932). In mimetite there are two lead positions, one fourfold (Pb_I) and one sixfold (Pb_{II}). A comparison between the two structures shows, that the fourfold lead position of mimetite corresponds to two lead atoms (Pb_{II}) and two chlorine atoms in finnemanite. Thus in finnemanite the chlorine atoms have replaced two lead atoms, which in their turn have changed position with chlorine.

The x - and y -parameters of the sixfold lead position of finnemanite and mimetite differ considerably and the coordination around these lead atoms is quite different. In mimetite there is a ninefold coordination with ionic $Pb-O$ bonds, while in finnemanite there is only fourfold coordination with bonds of mainly covalent character.

Arsenic occupies a sixfold position in both of the structures but with very different parameters. In mimetite the pentavalent arsenic atom is surrounded by four oxygen atoms in tetrahedral configuration, while in finnemanite there is a pyramidal arrangement of oxygen atoms around the trivalent arsenic.

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The paragenesis of the Varuträsk pegmatite

Arkiv för Mineralogi och Geologi
Band 2 № 2

Corrections.

- page 10. Plate IV (illustrations. line 10) read VI.
» 14. through (line 17 from bottom of page) » trough
» 15. Fig. 1. Boden » Boliden
» 116. delete reference (line 10 from top) (25,p. 459)

Plate II. Occurrence of varulite (if not corrected) should be removed from column for the lithium unit to that of the sodium unit.

Varulite, Tourmaline group, Spodumene, Mica group, Cookeite, Petalite, Pollucite, Albite, Microcline perthite

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Figs. 39, 40, 42, 52, 57, and 62 are reproduced from O. Adamson's paper on the feldspar group (Minerals of the Varuträsk Pegmatite XXXI, G.F.F. 64, p. 311).

Figs. 6, 7, 8, 9, 10, 15, 16, 17, 18, 19, 27, 28, 29, 33, 34, 45, 61, and 65 are reproduced from the original descriptions of the minerals in the series Minerals of the Varuträsk Pegmatite I-XXXV (G.F.F. 59-67, 1937-1945).

The remaining 45 figures have not been published previously.

Abbreviations

G.F.F.—Geologiska Föreningens Förhandlingar (Stockholm).

Arkiv för kemi etc.—Arkiv för kemi, mineralogi och geologi. Kungl. Vetenskapsakademien, Stockholm.

Arkiv för mineralogi—Arkiv för mineralogi och geologi. Kungl. Vetenskapsakademien, Stockholm. (New series beginning 1950.)

All other abbreviations are given in generally accepted terms.

The capital letters B to T on the map, Pl. III, indicate localities or environments referred to in the text.

I. Introduction

During the years 1937 to 1946 a series of papers under the common title "Minerals of the Varuträsk Pegmatite" have been published by members of the Mineralogical Department of the University of Stockholm. In these papers some thirty minerals or mineral groups, characteristic of the pegmatite, have been described. In most cases, however, only incidental reference was made to paragenetic questions connected with the minerals then under consideration. A more detailed account of the paragenetic development was instead planned to succeed the mineral descriptions, but was postponed on the chance that new minerals might be forthcoming during continued mining processes. Recent years have, however, not brought any new material to light, and as mining operations were successively reduced and now suspended, there is at present little hope of finding further matter of interest. An exposition of the conclusions which can be drawn regarding the paragenesis of the pegmatite will therefore now be attempted.

This publication was originally only intended to deal with the paragenesis of the pegmatite. For several reasons it has, however, seemed opportune to include a summary description of the characteristic minerals of the pegmatite, previously disconnectedly published in another journal (Mineral of the Varuträsk Pegmatite I-XXXVII, G.F.F., Vol. 59-68, 1937-1946). The principal reason for giving such a recapitulation is that in some cases a revision of former descriptions has been required and that new points of view have been commented on in other connections. A further motive is that it may be found useful to gather the essential features of the pegmatite in a collective publication.

In the first part of this paper a brief description of all earlier described minerals will therefore be given. The sequence, with exception of more insignificant alteration products, mentioned in connection with their parent mineral, will follow the Strunz tables.

The second part of the paper will deal with such observations which may elucidate the sequence of mineralisation in the pegmatite and offer conclusions regarding its paragenetic development.

Through the courtesy of the Boliden Mining Company I was given opportunity of visiting Varuträsk and making collections of the minerals during the course of many years. Dr. Erland Grip, now chief geologist of the company, has given his helpful cooperation to the preparation of the map which in its general contours is founded on the prospecting charts of the Boliden Co. He has also contributed much information while mining operations were still in progress and given every assistance in furthering the investigation of the pegmatite. Messrs. G. Gustavsson, B. Tornqvist, and A. Wirstam, mining engineers of the Boliden Company, have also contributed helpfully with sketch maps and photographs of some localities of special interest.

The identification and description of the many different minerals has been much facilitated by the generosity of the Boliden Company in letting their qualified chemist, Miss Thelma Berggren, analyse any specimen requiring chemical investigation. Of the 86 new analyses published in the series "Minerals of the Varuträsk Pegmatite" no less than 79 have been made by Miss Berggren. Many of these refer to minerals of complicated composition. Miss Berggren's efficient collaboration merits a special acknowledgement.

Several papers dealing with the earlier description of minerals or mineral groups from Varuträsk have been written independently by different co-workers in this Institute. I would like to mention especially the following persons who have taken an active part in identifying and describing some of the minerals which successively came to light. Professor Brian Mason, at that time Research Fellow of the University of New Zealand at this Institute, took up for renewed investigation the whole series of lithium-manganese phosphates and their alteration products. His paper was published under the title "Some iron-manganese phosphates and their alterations products with special reference to material from Varuträsk" (44). Dr. P. E. Wretblad undertook to re-investigate the mineral allemontite and was able to identify different phases of the mineral. His paper was published under the title "Die Allemontite und das System As-Sb" (76). Dr. Olge Adamson has described the minerals of the feldspar group, in which a great number of analyses and optical determinations are given (1). Miss Thelma Berggren has herself on several occasions visited the pegmatite. In more than one instance she was the first to collect specimens of a mineral not previously identified from the locality. She also undertook to analyse all the different varieties of the mica minerals and discuss their chemical composition. The results are published in two papers, entitled "Analyses of the mica minerals and their interpretation" and "Some new analyses of lithium-bearing mica minerals" (4, 5). Miss Brita Lundblad has determined the optical constants of all the analysed micas, published under the title "Optical studies of the analysed micas from Varuträsk" (40). Dr. O. Gabrielsson has in essential parts contributed to the description of the tourmaline group. In a map he has shown their distribution in the field and in many tables elucidated connections between chemical composition and optical constants (53). Professor A. Westgren (74, p. 232) and Professor O. Ödman (79) have co-operated in the description of the minerals stibiotantalite and microlite. Miss Karin Åmark has published an X-ray study of a stanniferous columbite from the pegmatite (78).

Dr. A. von Volborth has kindly undertaken to revise measurements of a columbite crystal and made a drawing of the same.

Professor O. Mellis of the Institute has taken most of the photographs for the paragenetic part of the mineral descriptions. Mr. E. G. French has kindly revised the English text.

The author wishes to express his sincere thanks to the Boliden Company for ready courtesy and financial support during the preparation of this paper. His cordial thanks are also due to Dr. Erland Grip and to other officials of the Company as well as to all collaborators mentioned above, for interested and helpful co-operation in different connections. Special thanks are due to Professor Brian Mason both for his efficient participation in the description of many of the rare phosphate minerals and for his critical revision of certain parts of the manuscript.

On account of the curtailed form in which most of the following mineral descriptions will be given, abbreviated from the original papers, it has not always been feasible in every case to acknowledge quotations from my collaborators. Reference will, however, be given under each mineral description which I hope may avert any reprimand for plagiarism on my part.

References not referring to mineral descriptions are assigned to a separate concluding list.

II. Location and general outline

The Varuträsk pegmatite was incidentally discovered in 1933. The owner of the ground had noticed an outcrop of quartz in an outlying forest clearing and put in a charge for speculative blasting. A fine-grained pink rock was thereby exposed. Dr. O. Baeckström, then chief geologist to the Boliden Mining Company, was shown a sample of this rock and recognised it as lepidolite. The occurrence of a lithium pegmatite on the site was thus established. Further blasting was then undertaken by the Boliden Company and shortly resulted in the discovery of other lithium minerals such as spodumene, petalite and amblygonite. As more methodical mining operations were begun, many other, and in some cases rare minerals were disclosed.

The Varuträsk pegmatite is situated 22 km SE of the Boliden gold mine and 15 km from Skellefteå town on the Baltic coast (Fig. 1). The bus line between Boliden and Skellefteå passes within one km of the quarries.

Regarding the surrounding formation of the pegmatite, Professor Gavelin, who has participated in the mapping of the district, has kindly given the following review:

“The Varuträsk pegmatite occurs in an amphibolitic rock, intercalated in an extensive schist formation, which has in this region been named the phyllite series on account of fine-grained sediments comprising a dominant part of the same. Amphibolitic rocks are very usual within the schist formation and may in different parts have originated from basic lavas or tuffs or from calcareous or marly sediments. It is not always possible in each case to determine from which rock formation the amphibolitic rocks have developed. The occurrence of cordierite, observed in amphibolite around the Varuträsk pegmatite, may signify that it represents, at least in part, a metamorphosed sedimentary rock (Fig. 2). In other parts it may have originated from basic lavas intercalated in the schist formation.”

The pegmatite is now in general assumed to be genetically connected with the so called Skellefte granite, representing fine-grained outcrops of the more widespread Revsund granite.

The pegmatite forms a through-like tabular body, striking NNE–SSW (Fig. 3). In the eastern wing the dip is about 30° WNW, whereas the western wing lies all but horizontal. The exposed outcrop is 350 m in length. The thickness varies from some three meters up to 30 meters. The elevation is 120 m above sea level.

The pegmatite is bounded both above and below by an amphibolitic rock of the same nature as that found in the surrounding rock complex. No obvious change of the amphibolite is manifested. In many places, however, xenoliths of amphibolite are found within the pegmatite and then distinct reaction zones are often to be observed. Along the contact an inner zone of small red garnets may then be seen, followed by a zone of albite and black tourmaline, the latter often in large individuals (Fig. 4). These contact phenomena indicate that epimagmatic conditions must then have been prevalent.

The lithium-bearing parts of the pegmatite occur in two separate lenses, intersected by a part of the pegmatite devoid of lithium minerals. The two lenses lie about 50 m apart.

Four stages in the mineralogical development of the Varuträsk pegmatite can be distinguished. The first stage is taken to represent the original zonal structure of the

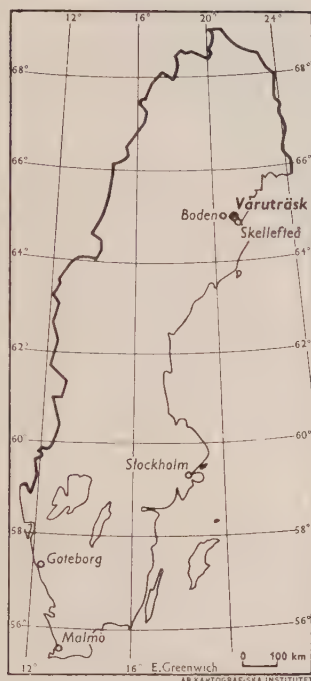


Fig. 1. Location of the Varuträsk pegmatite.

Fig. 2. Drill core from the hanging wall north of H₂ on the map. Offshoots of pegmatite in the surrounding amphibolite.
Photo E. Grip.



pegmatite body, formed by fractional crystallisation from the wall rock inwards. The second stage represents the different units of replacement. The third stage includes the phase of hypogene alterations. The supergene alterations are referred to a concluding fourth stage.

As all mining operations at Varuträsk have been suspended for the last four years, the quarries are now waterfilled and intervening parts to a great extent hidden by overgrowth. Many observations in the following paragenetic description must therefore refer to earlier records. It is possible that a more precise distribution of certain minerals might have been established if circumstances had permitted a renewed detailed supervision. Most observations of significance for the paragenetic description have, however, in different ways been able to be verified.

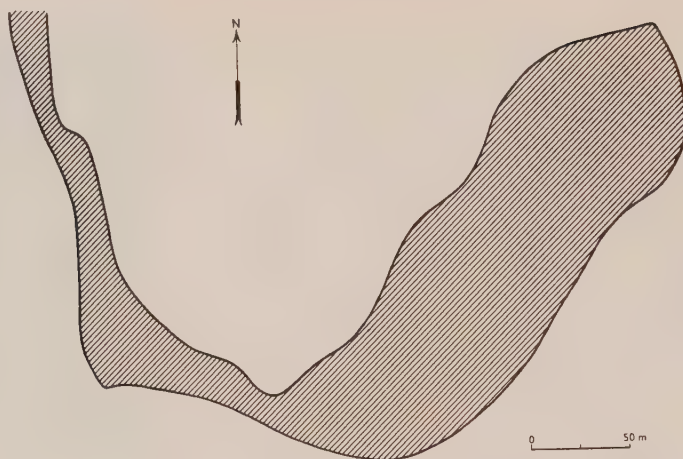


Fig. 3. Surface extension of the Varuträsk pegmatite. Scale 1:1330.

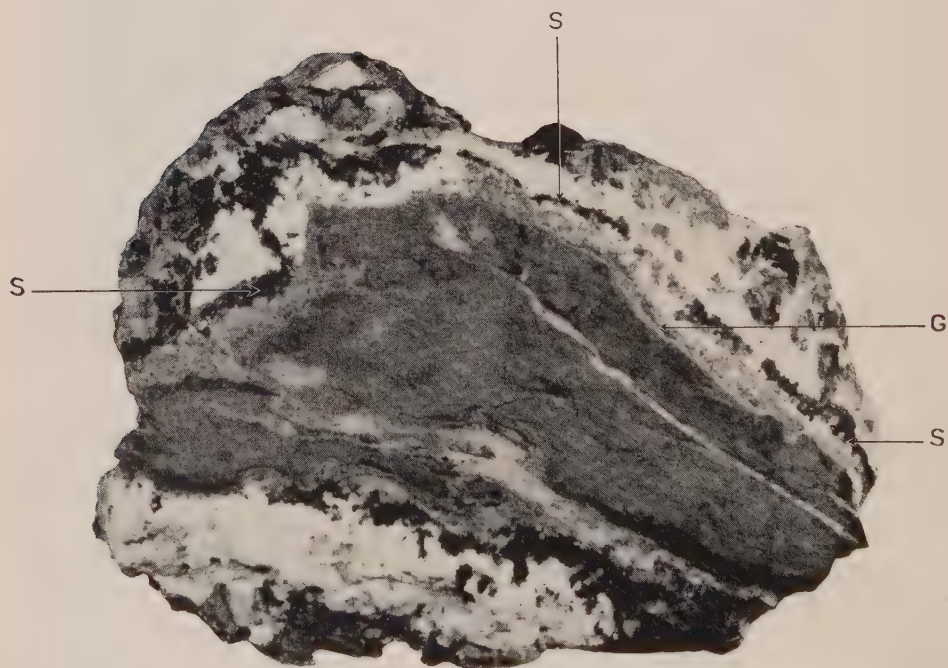


Fig. 4. Xenolith of amphibolite within the wall zone, showing an inner reaction zone of brown garnet (G) and an outer zone of schorlite (S). $\times \frac{1}{4}$. Photo S. Hedlund.

III. Summary of former mineral descriptions

The recapitulation of earlier descriptions of the minerals of the Varuträsk pegmatite will in the following pages vary in extent; minerals of more usual occurrence will be treated summarily. The original analyses as well as optical data will, however, in all cases be reconsidered.

Minerals presenting features of special interest will on the other hand be given more detailed attention. This will also be the case when in additional papers or in the form of personal communications new aspects have been brought to light concerning a certain mineral. Descriptions to which these conditions apply are: the allemontite group, columbite, stibiotantalite, microlite, arsenostibite, the lithium-manganese phosphates (triphyllite and lithiophilite) and their alteration products, varulite and its alteration products, manganvoelckerite and the mica minerals.

Of these minerals Dr. Wretblad has himself summarized the description of the allemontite group. Professor Mason has likewise summarised the description of the alkali-manganese phosphates and included new aspects on the origin of varulite. He has also given the results of a complementary investigation of the mineral arsenostibite. Professor Wickman has undertaken a new investigation of manganvoelckerite with regard to its validity as a definite mineral species.

1. Allemontite

The occurrence of allemontite in a pegmatite was unexpected as the mineral had previously been found only in ore deposits. Haidinger named the mineral in 1845, when describing it from the original locality at Chalanches near Allemont.

The occurrence at Varuträsk is restricted to a small prospecting pit, hardly two meters in diameter. In insignificant quantities the mineral also occurs dispersed in the adjacent rock. Though mining operations were continued in the vicinity, no further find of the mineral has been made. In the prospecting pit two large samples were obtained, weighing 490 and 370 g respectively. These are now in the museum collection of the Mineralogical Department of the University of Stockholm and of the Natural History Museum in Stockholm.

The allemontite is in the field associated with cleavelandite, green tourmaline and dark vitreous quartz. It is tin-white in colour. The larger samples (type I below) show well-defined cleavages parallel (0001). Fig. 5.

The first description of allemontite from Varuträsk was published in 1937 (P. Quensel 1937, K. Ahlborg and A. Westgren 1937). In 1941 Wretblad undertook a renewed investigation of the mineral. New aspects of importance thereby came to light (P. E. Wretblad 1941). Dr. Wretblad has kindly summarized the results of his work as follows:

“Allemontite is not a name for a specific mineral. Investigations of material from Varuträsk have made clear that three minerals occur, two of them being the native metals arsenic and antimony, in both cases containing a certain amount of the other metal. The third mineral has the approximate chemical composition AsSb. As this substance lacks a name, I in my earlier paper proposed the name stibarsen. The name wretbladite has later been proposed by Gagarin and Cuomo for this modification of the allemontite group (cf. *Am. Min.* 36, 1951, p. 638) but is not here applied.

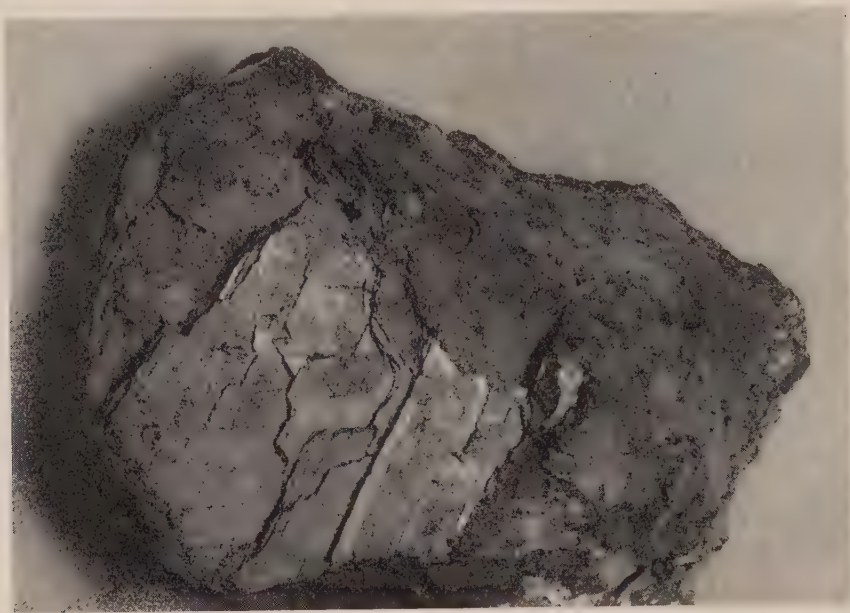


Fig. 5. Allemontite, heterogeneous species (type I). Nat. size. Excavation G₂. Photo O. Mellis.

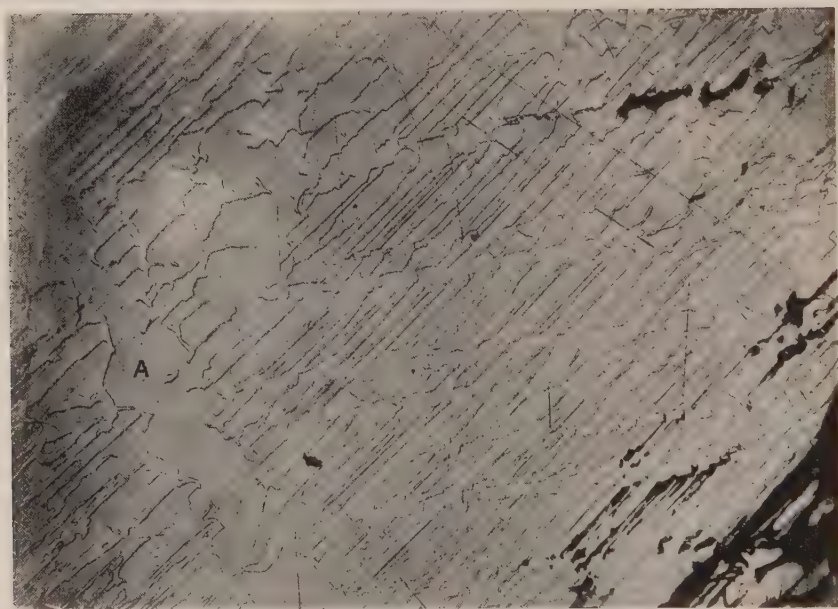


Fig. 6. Polished section of allemontite, type I. A = AsSb phase. $\times 10$.

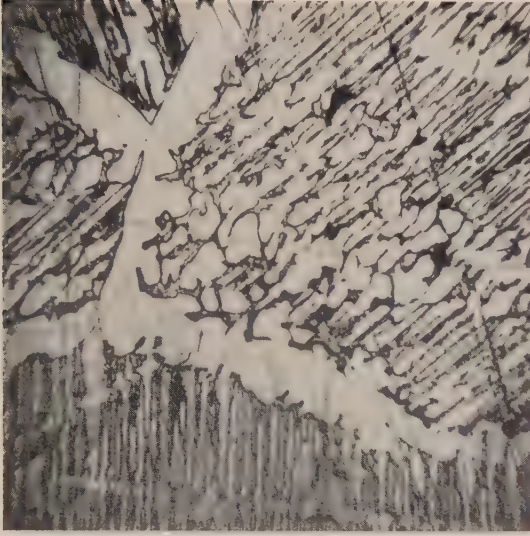


Fig. 7. Polished section of allemontite, type I. Etched with H_2O_2 . AsSb phase unattacked. Sb phase darkened. $\times 25$.

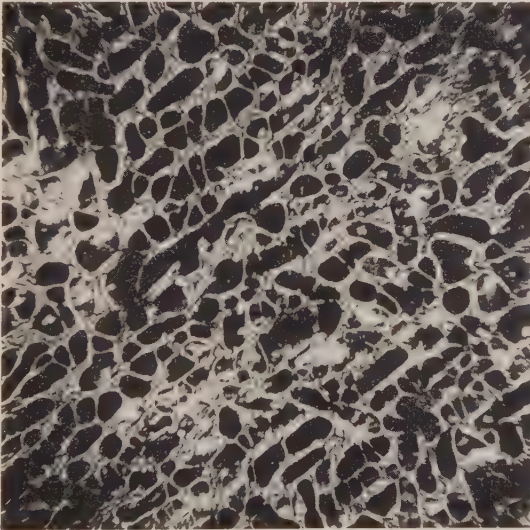


Fig. 8. Polished section of allemontite, type III. Etched with H_2O_2 . As phase unattacked. AsSb phase darkened. $\times 25$.

“The three different types of allemontite from Varuträsk have the following mineralogical composition:

Type I (heterogeneous) consists of stibarsen and native antimony.

Type II consists of homogeneous stibarsen (AsSb).

Type III (heterogeneous) consists of stibarsen and native arsenic.

"All these three modifications have a crystalline habit. Types II and III also occur with a gelable structure at Příbram (l.c. p. 24). Type I has first been identified from Varuträsk (later I have identified it also from Allemont, l.c. p. 34). Type II was also first found at Varuträsk (Quensel 1937). Type III was first found at Allemont in 1772 (Rammelsberg 1844). No new find of this type has been recorded until I was able to identify it from Varuträsk.

"In order to distinguish the different types and the minerals therein it proved necessary to use etch-reactions on polished surface, and in my paper a number of such reactions were described (l.c. p. 28).

"The heterogeneous types are most abundant. When the investigation had shown that these consisted of two different phases, an Sb-rich type I and an As-rich type III, it was established that the difference between them was great enough to distinguish them with the naked eye. Usually not even a polished section is necessary, as the air-etching gives sufficient contrast between the integrating minerals. Arsenic gets a darker colour than stibarsen which in its turn gets darker than native antimony.

"Type I shows macroscopically an appearance reminiscent of Widmanstätten structure (Fig. 6). A visible coarse network is microscopically found to be homogeneous. It consists of AsSb (stibarsen). The meshes of the network are filled with a very regular graphic accretion of AsSb and Sb, usually with each phase uniformly orientated over large areas (Fig. 7).

"In the type II rounded grains can be observed with the naked eye (Fig. 8). They consist of AsSb. Under the microscope the substance between the grains as a rule is found to consist of AsSb and As in eutectoid proportions. The grains may macroscopically be suggestive of homogeneous crystals but a microscopic examination reveals that every crystal of AsSb contains a certain amount of the interjacent substance.

"The allemontite type I and III have been identified from Allemont. Both differ somewhat in appearance from the Varuträsk types, elucidated in my former description (l.c. p. 36).

"In apparent contrast to these mineralogical facts, artificially prepared AsSb-alloys form a continuous homogeneous series in any proportions. Some experimental syntheses, described in my paper, have led me to the assumption that the homogeneous alloys on each side of the composition AsSb principally do not represent states of equilibrium but consist in under-cooled solid solutions, which do not decompose in reasonable time. In native AsSb-alloys the decomposition is accelerated by the impurities, acting as germs of crystallisation. Only in the neighbourhood of the composition AsSb the homogeneous phase represents a true equilibrium at ordinary temperature, and constitutes the allemontite type II. With essentially less than 50 atomic % As the allemontite forms type I (Sb + AsSb), and with essentially more than 50 atomic % As it forms type III (As + AsSb). The everpresent component in all modifications of allemontite will be stibarsen, AsSb."

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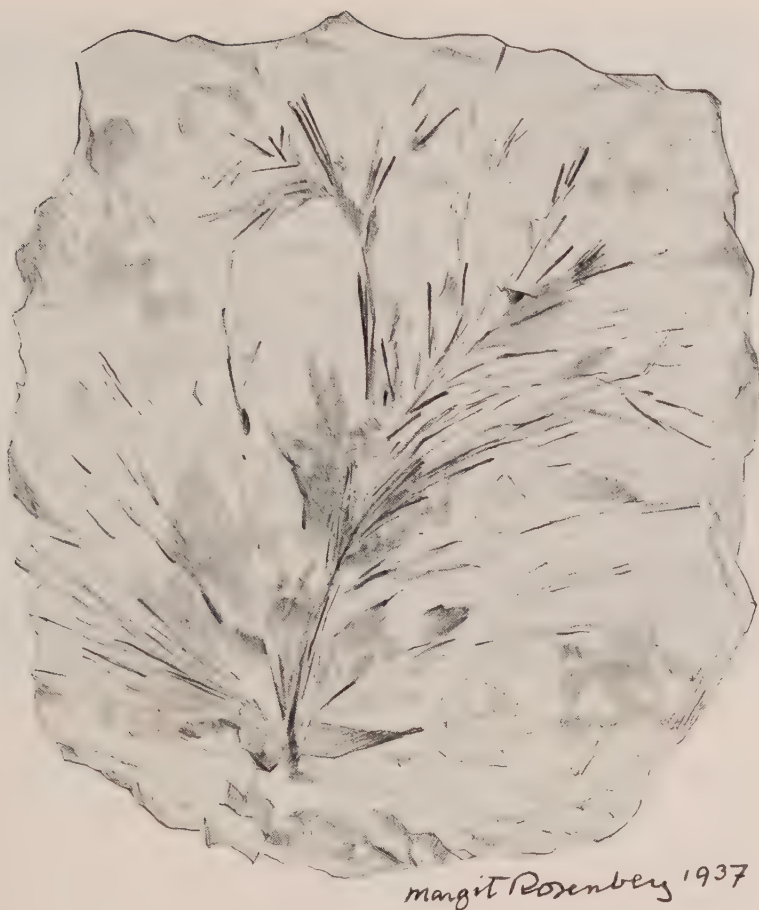


Fig. 9. Sheaf-formed löllingite. Excavation near T. $\times \frac{3}{4}$.

2. Löllingite

Löllingite has only been found in two localities at Varuträsk. The richest find is close to the hanging wall east of H_2 , where the mineral occurs in a lens of quartz, muscovite and black tourmaline pertaining to the wall zone assemblage. It is there spread in thin, bladed grains, 1–2 cm in length. In polished sections one often finds that the grains are corroded by the surrounding minerals, mainly quartz. Tiny specks of native bismuth can be observed on the sutures of the adjacent minerals.

Löllingite of a different appearance is found in the small prospecting excavation T in the eastern wing. It is there developed in radiating sheaves up to 10 cm in length, associated with quartz, muscovite and later invaded by cleavelandite (Fig. 9).

Two analyses have been made by Miss Berggren from the latter occurrence, given below.

Table 1

	1	2
As	71.7	68.58
Fe	27.2	28.78
S	0.8	1.64
Sb	—	0.00
Bi	—	< 0.01
Co	—	0.00
Cu	—	0.00
insoluble	0.8	1.00
	100.5	100.01
Sp.gr.		7.071

- 1: Löllingite, excavation 50 m east of quarry P.
2: Löllingite in boulder close by locality for analysis No. 1.

Löllingite has long been known to occur in lithium pegmatites from different localities. Its participation in the mineral assemblage of the Varuträsk pegmatite was therefore in no way unexpected.

REFERENCE

ÖDMAN, O. (1942), Minerals of the Varuträsk Pegmatite XXXIII. G.F.F. 64, p. 277.

3. Fluorite

Fluorite has not previously been described from Varuträsk, where it is of rare occurrence. The few samples, which have been found, occur in small vugs together with dark vitreous quartz. Its rare occurrence indicates that a deficiency of halides in the mineralising solutions has prevailed during the stage of its deposition as also during previous stages of mineralisation, in the pegmatite. This circumstance is commented on under a following heading (cf. p. 55).

The colour of the mineral is pure white. It is non-luminiscent. Crystal faces are defectively developed in the form of fractured cubes.

An analysis by Miss Berggren gave the following result:

Table 2.

Ca.	50.99	Li ₂ O.	0.00
F	48.36	Na ₂ O	0.00
SiO ₂	0.02	K ₂ O	0.00
TiO ₂	0.00	SO ₃	0.00
Al ₂ O ₃	0.03	P ₂ O ₅	0.00
Fe ₂ O ₃	0.09	CO ₂	0.00
BaO	0.00	H ₂ O ⁺	0.20
SrO	0.001*	H ₂ O ⁻	0.00
MgO	0.05		99.74

* spectogr. determination

4. Quartz

Quartz participates throughout the whole period of mineralisation of the Varuträsk pegmatite. In the core it occurs as milky quartz in large masses. In later stages of

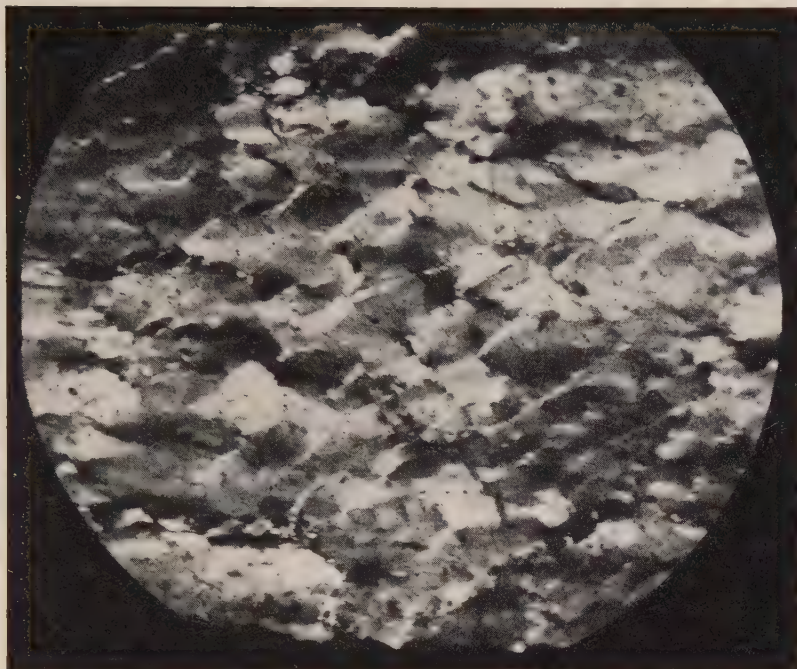


Fig. 10. Strain structure in core quartz. Quarry P. Nicols crossed. $\times 12.5$.

mineral deposition it is more universally developed as vitreous quartz, in colour varying from colourless to smoky or deep black.

Idiomorphic forms are all but absent. During the whole period of mining, developed crystal faces have only been found in one small vug. The larger crystals there have a distinct platy habitus parallel the prism (1010).

The milky quartz of the core often shows a beautifully developed strain structure. Under crossed nicols a mosaic of curved ridges are seen intersecting each other at acute angles, extinguishing almost, but not exactly, uniformly (Fig. 10).

REFERENCE

QUENSEL, P. (1942), Minerals of the Varuträsk pegmatite XXXIV. G.F.F. 64, p. 283.

5. Cassiterite

Cassiterite is not uncommon in the Varuträsk pegmatite, though dispersed in small amounts, generally associated with cleavelandite. In some localities it can occur in relatively large individuals, in others it forms strings of small grains (Fig. 11). Except in radiating crystals from such strings, crystal faces are not developed or are only faintly indicated.

The cassiterite is pitch black in colour. In thin section it is reddish-brown.

Two analyses by Miss Berggren are given in Table 3. Analysis 1 is of a specimen from the quarry P, analysis 2 from the prospecting pit G₁.



Fig. 11. Strings of cassiterite in cleavelandite. To the right gray lepidolite. Excavation G₁. $\times \frac{2}{3}$. Photo O. Mellis.

Table 3.

	1	2
SnO ₂	99.30	98.80
Nb ₂ O ₅		
Ta ₂ O ₅	0.24	0.76
FeO	0.55	0.42
MnO	0.01	0.06
H ₂ O ⁺	0.09	0.13
H ₂ O ⁻	0.00	0.00
	100.19	100.17

REFERENCE

QUENSEL, P. (1941), Minerals of the Varuträsk Pegmatite XXX. G.F.F. 63 p. 300.

6. Columbite

Columbite is not uncommon at Varuträsk though generally only found in small platy individuals between the cleavage planes of coarse-grained cleavelandite. More irregular masses of small dimensions occasionally occur. The platy crystals may attain a length of 2–3 cm.

One single crystal of larger size has however been found and displayed many faces (Fig. 12). A. von Volborth kindly undertook to measure the crystal and identified the following forms:

b (010), *a* (100), *g* (110), *m* (130), *k* (011), *e* (201), *o* (131), ? (121), *u* (111), *n* (211). (Indices according to Goldschmidt.)

A drawing of the crystal by Volborth is pictured in Fig. 13.¹

¹ The deduced forms by O. Lundquist in the original publication have been found erroneous and must be substituted by those now given.

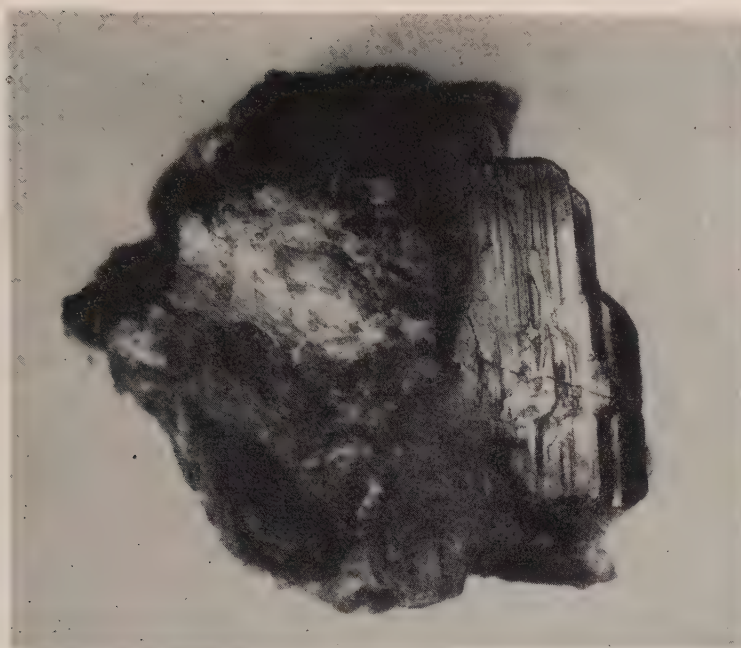


Fig. 12. Crystal of columbite. Nat. size. Photo O. Mellis.

Two analyses of the platy crystals by Miss Berggren are given in Table 4, Nos. 1 and 2.

Table 4.

	1	2	3	4
Nb ₂ O ₅	61.30	61.00	35.90	31.83
Ta ₂ O ₅	13.60	17.80	17.98	21.46
Al ₂ O ₃	1.20	—	—	—
TiO ₂	1.07	0.76	0.14	0.10
SnO ₂	3.14	0.76	32.68	32.65
SiO ₂	1.16	0.00	—	—
FeO	10.70	5.62	0.81	0.55
MnO	8.52	13.50	12.00	12.81
CaO	—	—	0.00	—
MgO	—	—	0.10	—
Na ₂ O	—	—	0.15	—
H ₂ O ⁺	—	—	0.09	0.11
H ₂ O ⁻	—	—	0.25	0.35
	100.69	99.44	100.10	99.86
Sp. gr.	5.623	5.460		

The two analyses show marked differences in the proportions between MnO and FeO. In the first analysis Mn:Fe is 6.6:8.3 whereas in the second analysis Mn:Fe is 10.5:4. Though the content of Mn proportionally is so much higher in the second analysis, it is according to the proposal given in Dana's System of Mineralogy (1944, p. 783), not sufficient to name the species managanoan columbite as this name there is restricted to columbite with Mn:Fe > 3:1.

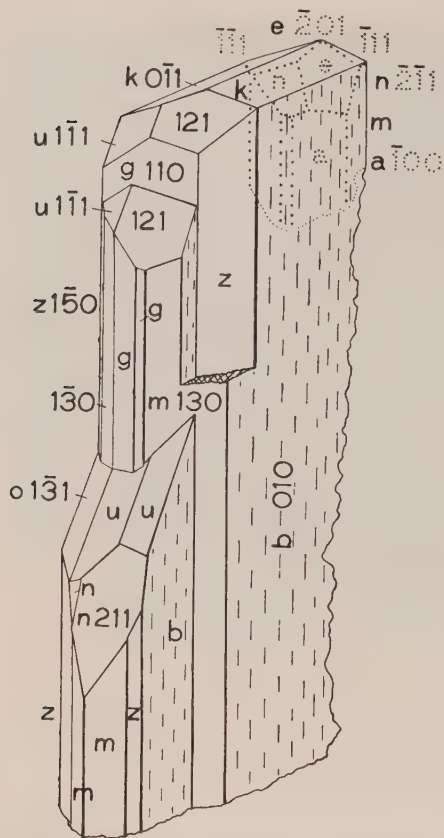


Fig. 13. Drawing by A. von Volborth of columbite crystal Fig. 12. (Indices according to Goldschmidt.) $\times 2\frac{1}{2}$.

Later some specimens of columbite were found which differed somewhat from the first analysed samples. The streak was lighter in colour than the brownish-black streak of the previous finds of the mineral. The analyses by Miss Berggren showed a content of over 32 % SnO_2 as seen in Table 4, nr. 3 and 4.

Miss Karin Åmark kindly undertook to make an X-ray investigation of the two analysed specimens. Powder photographs proved that in its present state the mineral represented a mechanical mixture of cassiterite and a columbite—tantallite phase. A study of polished sections by Ödman resulted in the same conclusion. He says: "All details of the microscopic structure indicated that even in its primary state the mineral has only been a purely mechanical mixture of cassiterite and a columbite phase."

The name stanniferous columbite, applied in the original publication, seems no longer to be justified and should in this connection be discarded.

REFERENCES

- QUENSEL, P. and BERGGREN, TH. (1938), Minerals of the Varuträsk Pegmatite XX. G.F.F. 60, p. 22.
 QUENSEL, P. (1941), Minerals of the Varuträsk Pegmatite XXX, G.F.F. 63, p. 301.
 ÅMARK, KARIN (1941), Minerals of the Varuträsk Pegmatite XXIX, G.F.F. 63, p. 295.
 ÖDMAN, O., see QUENSEL, P., 1941 p. 303.



Fig. 14. Manganotantalite (manganoan tantalite). Underground workings from the shaft. Nat. size. Photo O. Mellis.

7. Tantalite

During earlier stages of mining operations only a few specimens of tantalite came to light. Later, however, large quantities of the mineral were found underground around the shaft (Fig. 14).

Table 5.

	1	2
Nb ₂ O ₅	3.00	25.68
Ta ₂ O ₅	74.00	56.86
FeO	9.81	0.20
MnO	10.79	15.95
CaO	0.00	0.00
MgO	0.00	0.31
Na ₂ O	—	0.24
K ₂ O	—	trace
SiO ₂	1.14	—
SnO ₂	0.00	0.19
TiO ₂	0.73	0.12
H ₂ O ⁺	—	0.17
H ₂ O ⁻	—	0.21
	99.47	99.93
Sp. gr.		6.349

1: Tantalite, find 1935.

2: Manganotantalite (manganoan tantalite), find 1940.

The tantalite is brownish black, in thin edges brown and then translucent. In thin sections it has a mustard-yellow colour. Twinning after *c* (021) is often developed.

Two samples of the mineral have been analysed by Miss Berggren, given in Table 5.

According to the proposal in Dana's System of Mineralogy (1944, I, p. 783) the name manganoan tantalite (manganotantalite) should be restricted to such species where Mn:Fe > 3:1. The name manganoan tantalite is in this respect fully justified for the analysed specimen No. 2, where Mn:Fe is 12.35:0.155.

This analysis is extreme in its low content of FeO and high content of MnO. Of all published analyses of tantalites only two can compete in this respect. These are from Branchville, described by Comstock, and from Mt. Apatite, described by Schaller, with 0.43 and 0.16 % FeO and 15.58 and 14.49 % MnO respectively (W. J. Comstock 1880, W. T. Schaller 1907).

REFERENCES

- COMSTOCK, W. J. (1880), Am. J. of Science XIX, p. 131.
 SCHALLER, W. T. (1907), Am. J. of Science XXIV, p. 154.
 QUENSEL, P. (1938), Minerals of the Varuträsk Pegmatite XI G.F.F. 60, p. 223.
 QUENSEL, P. (1941), Minerals of the Varuträsk Pegmatite XXIV, G.F.F. 63, p. 176.

8. Stibiotantalite

When visiting the Varuträsk pegmatite in 1936 some small specimens of a grayish semi-metallic mineral were found, subsequently identified as stibiotantalite. In 1939 a nodule containing in all around 12 kg of the mineral was excavated. Unfortunately most of it was shattered during excavation. One large sample, weighing 1.5 kg was, however, recovered. During renewed search the following years only some few

Table 6. Stibiotantalite-microlite compound.

	1	2	3
Nb ₂ O ₅	11.8	10.83	7.52
Ta ₂ O ₅	52.3	44.27	54.20
Sb ₂ O ₃ (insoluble)	24.92	21.28	14.63
Sb (soluble in HCl)	0.32	13.40	10.80
Bi ₂ O ₃	0.10	0.13	0.02
Al ₂ O ₃	0.50	trace	0.06
Fe ₂ O ₃	0.26	trace	0.32
TiO ₂	0.00	0.00	0.00
SiO ₂	1.38	0.60	1.36
MnO	0.08	—	1.28
CaO	5.32	6.12	7.38
MgO	0.00	0.00	trace
Na ₂ O	1.50	1.55	0.90
K ₂ O	0.00	0.19	0.11
H ₂ O ⁺	1.11	1.31	1.20
H ₂ O ⁻	0.16	0.07	0.06
	99.75	99.75	99.84
Sp. gr.	5.735		

- 1: Stibiotantalite, Varuträsk. Find of 1936.
 2: " " Find of 1939.
 3: " " Find of 1940.

further fragments of the mineral could be collected. All finds were restricted to two small prospecting pits around G_2 , close to the find of allemontite.

The stibiotantalite occurs in thick crystalline masses. Under the microscope one can clearly observe three phases present (Fig. 15). One is light yellow to colourless. The birefringence is extreme. A second phase, surrounding kernels of the first phase, has almost identically the same colour but it is isotropic. Both have all but identical high refractive indices. Without crossed nicols the two phases are hardly distinguishable from one another. The third phase was taken to be native antimony.

Three analyses by Miss Berggren are given in Table 6. Besides the main constituents (Sb_2O_3 , Nb_2O_5 , Ta_2O_5), a noteworthy content of CaO is present in all three analyses.

Professor A. Westgren and Mr. O. Rosén kindly undertook to make an X-ray investigation of the compound mineral and found that four phases were present. Though mainly a mixture of stibiotantalite and microlite, small amounts of native antimony and senarmontite were also identified. The results of their research are commented on as follows:

"The specimen in its present state is mainly a mixture of stibiotantalite and a microlite phase. The lattice dimensions of the former are the same as those of the Californian stibiotantalite, viz. $a = 4.92 \text{ \AA}$, $b = 5.54 \text{ \AA}$, $c = 11.78 \text{ \AA}$. The lattice constant of the microlite phase has been determined to be $a = 10.40 \text{ \AA}$. The previous anticipation that some native antimony is present in the specimen has also been verified. The strongest interferences in the X-ray pattern of Sb appear in the photographs of the mineral mixture. The lattice dimensions of this third phase were found to be normal, viz. with an edge of the face-centred unit rhombohedron of 6.226 \AA and an angle of $87^\circ 24'$. When the three phases had been identified, there remained, however, a few lines in the photographs, originating from something else. They were soon recognized as the strongest lines of senarmontite, proving that the specimen has a slight admixture of this mineral too. Its lattice constant was found to be $a = 11.13 \text{ \AA}$, in close agreement with the value 11.127 \AA , previously found in this Institute for pure cubic Sb_2O_3 . Although the stibiotantalite has the lowest symmetry of the four phases of the mineral mixture, a great multitude of its lines are visible in the photographs and several of them have a rather high intensity. The interferences of the microlite phase are also rather strong. These two substances are evidently the dominating constituents. The presence of Sb and Sb_2O_3 is indicated in the photographs only by their strongest lines and thus enter into the specimen in comparatively small quantities. The lines of these phases are further somewhat blurred and diffuse, which is an indication that they are formed secondary under poor crystallisation conditions." (O. Rosén and A. Westgren 1938.)

In order to try and elucidate the connection between the four phases, identified by the X-ray examination, Professor O. Ödman kindly undertook to examine some polished sections. The following is, with some abridgement, cited from Ödman's narrative:

"In all sections studied, the presence of the same components as previously found spectographically, namely stibiotantalite, microlite, antimony and senarmontite, has been established, although their amounts show great variations from specimen to specimen as is also evident from the analyses. As represented in Fig. 15, the stibiotantalite kernels are well developed, but in many

instances very little remains of this mineral and the largest parts of these areas, enclosed in the antimony network, are occupied by the microlite phase. The latter is developed all around the antimony veinlets.

"In the material from 1939 and 1940 the content of native antimony is rather high, as may be seen from the shining surfaces of the substance, which are covered with thin sheets of the metal. The analyses show 13.40 and 10.80 % Sb respectively.

"The network of the antimony veinlets follows mainly three directions at approximately right angles to each other. One direction is very prominent and it evidently follows cleavage planes in the stibiotantalite. The antimony veinlets are here rather narrow (on the average 0.01–0.005 mm) and are very sharply bounded. In the stibiotantalite the cleavage appears as distinct fissures in the polished surface.

"Two other systems of antimony veinlets are more irregular and attain a width of about 0.5 mm. They often have an intermittent or a winding course and may occasionally join in large areas, where the surrounding microlite has been considerably replaced. Replacement is noticed also along the walls of the veinlets and they may contain inclusions of microlite. In some cases extremely fine-grained and cloudy formations of antimony are seen in the microlite; they may even have dendritic forms.

"Under the microscope, with nicols crossed, the antimony is seen to be built up of small rounded, individual grains. In the antimony veinlets there have also been recognized stibnite and some gangue minerals. The stibnite is rare and occurs in small grains in some of the antimony veinlets. In a few cases stibnite in fairly large amounts was noted in veins or in patches with quartz, a micaceous mineral and antimony. A narrow fringe of the latter mineral separates the stibnite from the surrounding gangue minerals. The stibnite has also been seen to be cut by veinlets of antimony. In one section stibnite formed separate veinlets in the microlite; hardly any antimony was then present." (O. Ödman 1941.)

The study of polished sections has led Ödman to the following explanation of the intermixture of components in the mineral in question.

"The stibiotantalite, occupying the kernels in the network, constituted the primary mineral. An alteration of this mineral led to the formation of microlite and native antimony. A comparison between the composition of stibiotantalite and microlite indicates that such an alteration would involve an addition of CaO , Na_2O and H_2O . It is now believed that residual solutions entered the stibiotantalite along cleavage planes or fractures and caused it to break down. The antimony was set free and deposited on the fractures; some of it was combined with sulphur to form stibnite". (O. Ödman 1941, pp. 290–293.)

The chemical analyses as well as the X-ray investigation and the examination of the polished sections all agree that the two principle phases present are stibiotantalite and microlite. Native antimony is evidently present in very varying amounts and senarmontite in subordinate quantity. Taking the formula for stibiotantalite as $\text{Sb}(\text{Ta}, \text{Nb})\text{O}_4$ and for microlite $\text{Ca}, \text{Na}(\text{Ta}, \text{Nb})_2\text{O}_6\text{OH}$, Miss Berggren has calculated the percentage of the three components stibiotantalite, microlite and native Sb in the three analysed samples. The calculation gave the following results:

Table 7.

	Analysis 1	Analysis 2	Analysis 3
stibiotantalite	31.6	0.5	11.8
microlite	66.3	79.6	59.1
native Sb	2.1	19.9	29.1

The content of the three constituents are seen to vary proportionally in the three re-calculated analyses. In No. 2 practically all stibiotantalite has been altered to microlite and native Sb.

Ödman's explanation that the mineral compound represents successive alterations of stibiotantalite to microlite and native antimony is in this case unquestionable. In the first paper, describing the mineral compound (P. Quensel and Th. Berggren 1938), the microlite phase was assumed to represent the primary constituent. As a certain amount of antimony was assumed to have integrated in this phase, the name stibiomicrolite was suggested. In this case, however, the name is not justified as the variable amounts of antimony presumably emanate from decomposed stibiotantalite in the form of native Sb. In another connection the name stibiomicrolite (antimonian microlite) may still be legitimate (cf. p. 36 under the following heading).

REFERENCES

- QUENSEL, P. and BERGGREN, TH. (1938), Minerals of the Varuträsk Pegmatite XI. G.F.F. 60, p. 216.
 ROSÉN, O. and WESTGREN, A. (1938), *ibid.* XII. G.F.F. 60, p. 232.
 BERGGREN, TH. (1941), *ibid.* XXII. G.F.F. 63, p. 52.
 ÖDMAN, O. (1941), *ibid.* XXVIII. G.F.F. 63, p. 289.

9. Microlite

In the description of the foregoing mineral, microlite was observed to occur replacing stibiotantalite in varying amounts up to nearly 80 %. In this case, however, the microlite represents a secondary alteration product and was at the time for that description not known at Varuträsk in any other form.

When the Boliden Company in 1943 began sinking shaft in the eastern wing, a dark semi-metallic mineral was brought to light, subsequently identified as consisting mainly of microlite.

At first sight the mineral seemed pure black in colour. Even with a pocket lens, however, one could observe that the mineral in itself was light buff-coloured with a porcelain-like translucent lustre and that the black colouring was due to a metallic precipitation. In polished sections this was identified as a thin coating of metallic antimony.

In the few specimens available the mineral occurs as more or less rounded grains in petalite or in spodumene. The largest individual does not exceed 1 cm in diameter.

Under the microscope the mineral shows a somewhat patchy colouring from light yellowish-brown to all but colourless. It is fully isotropic throughout. In some sections indications of an octahedral cleavage can be observed.

An analysis of the mineral, made by Miss Berggren is given in Table 8:

In calculating the analysis the content of SiO_2 has been left out of consideration as due to impurity in the form of quartz veinlets, apparent in Fig. 16.

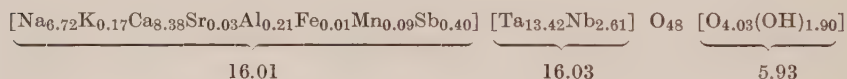
Dr. Byström kindly took some X-ray powder photographs of the mineral from which he could conclude that it has a lattice of pyrochlore type with $a = 10.40 \text{ \AA}$.

Table 8.

		Mol. ratios	Atomitic ratios
Nb ₂ O ₅	8.45	31.788	64
Ta ₂ O ₅	71.94	162.849	336
Sb ₂ O ₃ (+ Ce, U)	1.40	4.802	10
Al ₂ O ₃	0.27	2.649	5
Fe ₂ O ₃	0.03	0.188	0.4
TiO ₂	0.00	—	—
SiO ₂	0.95	—	—
FeO	0.00	—	—
MnO	0.16	2.256	2
CaO	11.38	202.924	203
SrO	0.08	0.772	1
MgO	0.00	—	—
Na ₂ O	5.04	81.298	163
K ₂ O	0.19	2.017	4
F	0.00	—	—
H ₂ O ⁺	0.42	—	—
	100.31		
Sp. gr.	5.619		

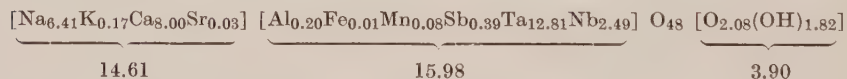
Professor Westgren has calculated the density under two different assumptions as follows:

"I. The pentavalent Ta and Nb atoms may be supposed exclusively to occupy the 16(*d*) position of the space group O_h^7 , while all the other metal atoms are situated in 16(*c*). The O atoms occupy 48(*f*) and together with OH ions also 8(*a*). The points of the 8(*a*) position are supposed to be partially vacant. The composition of the mineral would then in accordance with the analytical data correspond to the formula



and the density would be 6.02.

"II. The Fe, Mn and Sb ions may be supposed to be of higher valency and together with the Ta, Nb and Al atoms to occupy the position 16(*d*) while only Na, K, Ca and Sr atoms are situated in 16(*c*) which is partially vacant. The formula would then be



The density is in that case calculated to be 5.76.

"The latter of these two assumptions seems by far to be the most probable as the density was experimentally found to be 5.62."

Professor Ödman kindly undertook a metallographic analysis of the microlite in question and put at my disposal the following result:

"The polished section (Fig. 16) is chiefly composed of a relatively dark mineral which according to the X-ray analysis is microlite. Especially towards the borders of the microlite some remnants of a lighter gray mineral are to be

observed, more or less invaded by microlite (Fig. 17). According to X-ray photographs this mineral is stibiotantalite. Native antimony occurs in grains in the microlite as well as in cracks and veinlets in the same. In the surrounding spodumene larger grains of native antimony are found". (Minerals of the Varuträsk Pegmatite XXXV, G.F.F. 67, 1945, p. 20.)

In other samples obtained later, a more complex association is found. The mineral is in this case black in itself and not merely darkened by a superficial precipitation of native antimony. In outward appearance it much resembles samples of stibiotantalite.

Specimens of this type were found to consist of a mixture of manganotantalite and microlite with residual shedded fragments of stibiotantalite (Fig. 18).

An X-ray analysis by Byström defined the three phases as a columbite phase with $a = 5.09$ Å, $b = 14.39$ Å and $c = 5.76$ Å, a pyrochlore phase with $a = 10.41$ Å and stibiotantalite with the cell dimensions $a = 4.92$, $b = 5.54$ and $c = 11.78$ Å.

An analysis of the mineral compound by Miss Berggren is given in Table 9. In column 2 the mol. prop. have been recalculated after deduction of the tantalite phase, calculated on Mn as sole cation therein. Byström justifies this on the following account:

"For synthetic Mn-bearing compounds of columbite structure Karin Brandt found for MnTa_2O_6 $a = 5.081$, $b = 14.39$ and $c = 5.766$ Å. Comparing the values found for the columbite phase, it seems reasonable to assume that it mainly consists of Mn as cation." (Arkiv för Kemi etc. Bd. 17 A, Nr. 15.)

On this supposition Byström has calculated the composition of the pyrochlore phase (microlite) as $(\text{Ca.Na})_2(\text{Ta, Nb, Sb, Sn, Al, Fe})_2(\text{O, O, OH, F})_{6.94}$. The small surplus of 0.007 mol. prop. of Sb_2O_3 and $(\text{Ta, Nb})_2\text{O}_5$ would then be accounted for by the presence of some stibiotantalite. Byström reckons the proportions of the three phases to be about 40:20:1 (microlite:columbite:stibiotantalite). (Minerals of the Varuträsk Pegmatite XXXV, G.F.F. 1945, p. 24.)

Table 9.

	1	2		1	2
Nb_2O_5	8.63		FeO	—	
Ta_2O_5	60.23		MnO	4.90	
Sb_2O_3	7.61	0.1258	CaO	9.63	
Sb	2.23		SrO	0.05	0.1714
Bi_2O_3	0.00		Na_2O	2.36	
Al_2O_3	0.11		K_2O	0.24	0.0407
Fe_2O_3	0.37	0.0034	F	0.70	
TiO_2	0.00		H_2O^+	0.64	0.0723
SnO_2	1.26	0.0084			
UO_2	trace			99.75	
ThO_2	0.05		O for F	0.29	
SiO_2	0.74			99.46	
BeO	—				
MgO	trace				
Sp. gr.					5.902

1: Mineral complex of manganotantalite, microlite and stibiotantalite, Thelma Berggren anal. 1940

2: Mol. prop. after deducting manganotantalite, calculated on Mn as sole cation.

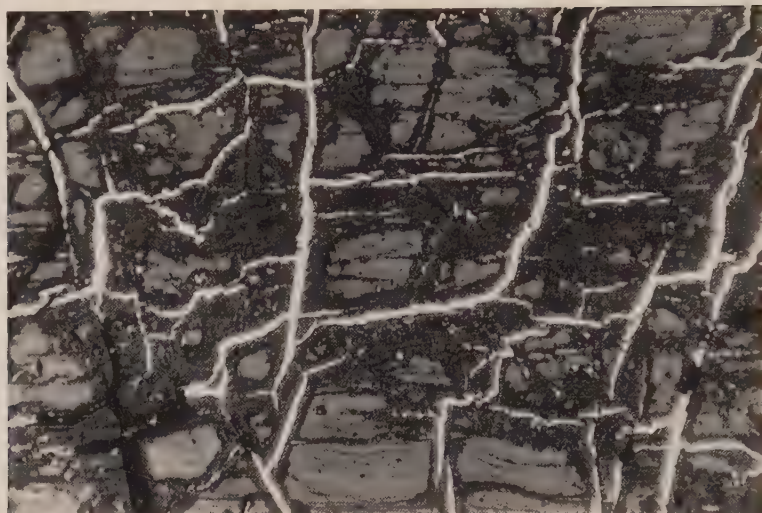


Fig. 15. Stibiotantalite (light gray kernels) surrounded by microlite (dark gray). White veinlets are native antimony. Black in veinlets is senarmonite. $\times 170$, ord. light. Photo O. Ödman.



Fig. 16. Microlite with specks of native antimony (white). In veinlet quartz (black) and stibiotantalite (light gray). $\times 90$. Photo O. Ödman.

In the polished section, (Fig. 18) taken by Ödman, one can clearly observe the relative proportions of the three participating constituents. In this case, however, the proportions between microlite and tantalite are other than those calculated by Byström on the basis of the analysis. In a mineral compound of this nature it is, however, to be anticipated that differences in this respect will occur.

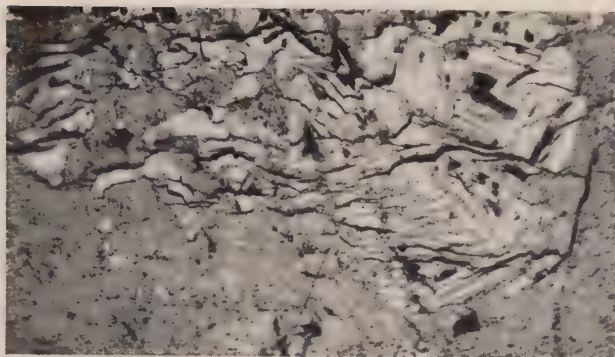


Fig. 17. Microlite (same occurrence as Fig. 16), here seen replacing stibiotantalite (light gray). $\times 90$. Photo O. Ödman.

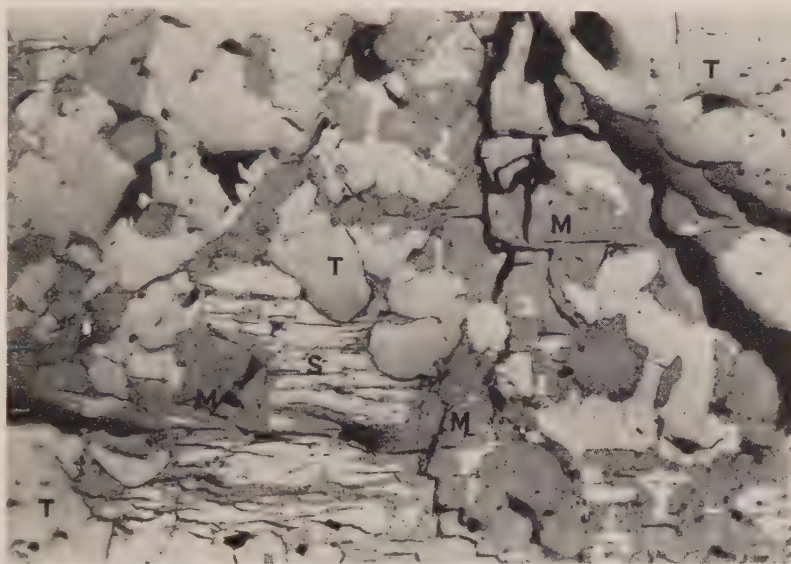


Fig. 18. Complex intergrowth, consisting of manganoan tantalite (T), microlite (antimonian microlite) (M.) with remnants of stibiotantalite (S). $\times 180$. Photo O. Ödman.

As noted in the previous description of stibiotantalite, it must now be considered established that the participating microlite phase there represents a replacement of stibiotantalite. In the polished section (Fig. 17) one can observe the same process in a minor scale. The residual stibiotantalite present is fringed by microlite.

The question now arises whether microlite, even when occurring in a pure state, as seen in Fig. 16, may not represent a complete replacement of stibiotantalite, there constituting the primary constituent. Several indications seem to point in this direction when the photographs here reproduced are compared. Fig. 15 in the preceding description of stibiotantalite would then indicate an initial stage where larger kernels of stibiotantalite are surrounded by areas of microlite. Fig. 17 above would represent

a somewhat more advanced stage, as is also the case in a minor scale in Fig. 18. Finally an almost complete replacement would be apparent in Fig. 16.

A further circumstance pointing in the same direction is that the precipitation of native antimony on the microlite, represented in Fig. 16, would find an explanation in the liberation of antimony in conjunction with the decomposition of stibiotantalite. In the analyses above, Miss Berggren determined the content of native Sb to be 2.23 % and of Sb_2O_3 to be 7.61 %. As Byström, calculating on this analysis, computed the proportions between the microlite-, columbite- and stibiotantalite phases to be 40:20:1, the amount of stibiotantalite must be insignificant.

If all the microlite present is taken to be a replacement of stibiotantalite as the primary constituent, most of the antimony thereof (theoretically 39.76 % Sb_2O_3 for the formula SbTaO_4) must have been dissolved and migrated. Of the remainder some fraction, represented in the analysis as 2.23 % Sb, may occur precipitated as native antimony, whereas the rest (7.61 Sb_2O_3 in the analysis), must be found in one of the two remaining components. As antimony, according to an analysis of manganotantalite from Varuträsk, is not present (0.00 % Sb) there remains microlite.

According to synthetical investigations, Rosén and Westgren have concluded that the Ca and Na atoms in microlite may be replaced by half their number of Sb^{III} atoms without any change of structure (l.c. p. 234). Even if in this case one cannot reckon with any definite percentage in connection with the analytical data, as the proportions of the integrating phases differ from one sample to another in the mineral compound, it is presumably admissible that a quantity of antimony can enter the microlite structure.

As other incidences, given above, seem to corroborate a supposition of the same nature, it seems justifiable to assume that the microlite at Varuträsk represents in all instances an alteration product of stibiotantalite and that during the replacement thereof a certain content of antimony has entered the microlite structure.

Since an antimony content is recorded in no former analyses of microlite, it may be assumed that the mineral in this specific instance represents an antimonian microlite to which the name stibiomicrolite was applied in the original description.

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ROSÉN, O. and WESTGREN, A. (1938), On the structure and composition of minerals belonging to the pyrochlore-atopite group and an X-ray analysis of disintegrated stibiomicrolite. Minerals of the Varuträsk Pegmatite XII. G.F.F. 60 p. 225.

10. Uraninite

Uraninite is of rare occurrence in the Varuträsk pegmatite and most specimens are of insignificant size. The mineral is most commonly developed in small well defined cubic crystals, rarely more than 5 mm in diameter. In a few specimens it occurs as larger irregular nodules and then in a more metamict state. In both cases the mineral is bordered by a zone of bright yellow oxidation products.

Material for analysis was taken from a crystal from which the yellow zone of oxidation could be removed. The material was, however, insufficient for determining stages of oxidation. Miss Berggren gives the following determinations:

Table 10.

PbO	16.64	Bi, Cu, Mn, S	0.00
U ₂ O ₈ (total U as U ₂ O ₈)	74.4	P ₂ O ₅	0.00
ThO ₂	1.12	SiO ₂	0.16
Fe ₂ O ₃	0.03	insoluble	0.32
Al ₂ O ₃	0.00	volatile	6.1
CaO	0.16		99.08
MgO	0.03		
He, N, H ₂ O, calculated as difference			0.92
			<hr/> 100.00

The analysis is remarkable with respect to the high content of PbO. Professor Wahl of the University of Helsingfors kindly undertook a determination of the lead isotopes with the following result:

Pb 206	81.94 %
Pb 207	15.26 %
Pb 208	2.80 %

An age determination thereon would give approximately 1700×10^6 years. Dr. S. Eklund has later calculated the age by the Rb/Sr method, conforming with a new half-life determination of Rb⁸⁷ (5.8×10^{10} years) (Eklund, S. 1946). He thereby arrived at a figure of 1740×10^6 years with a relative error of 12 %. The difference between Wahl's and Eklund's determinations hardly exceeds the limits of experimental error.

Some new determinations by others will be given in the summary of this paper and the results there be discussed (p. 122).

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11. Arsenostibite (*arsenian stibiconite*)

Summarized and extended by BRIAN MASON

"In 1937 Quensel described a yellow alteration product of allemontite consisting essentially of a hydrated arsenic-antimony oxide, and applied to it the name arsenostibite, which had been introduced by Adam (1869) (in the form arsenstibite) for a mineral of similar composition from Borneo. At the time of description it was thought that the Varuträsk material was non-crystalline, it being isotropic and to all evidence amorphous. However, Vitaliano and Mason (1952) have since made an X-ray investigation of arsenostibite and shown that it has the same structure as stibiconite and is evidently an arsenian variety of the mineral.

"Arsenostibite occurs at Varuträsk as a somewhat porous material with a prominent sulphur-yellow colour, coating or partly replacing allemontite. In many specimens one can macroscopically see how the individual crystals of allemontite have been more or less affected, the oxidation beginning along the cleavages and continuing until only small kernels of allemontite remain in the secondary product of alteration. A singular ladder-like structure is often produced (Quensel, 1937), the oxidation

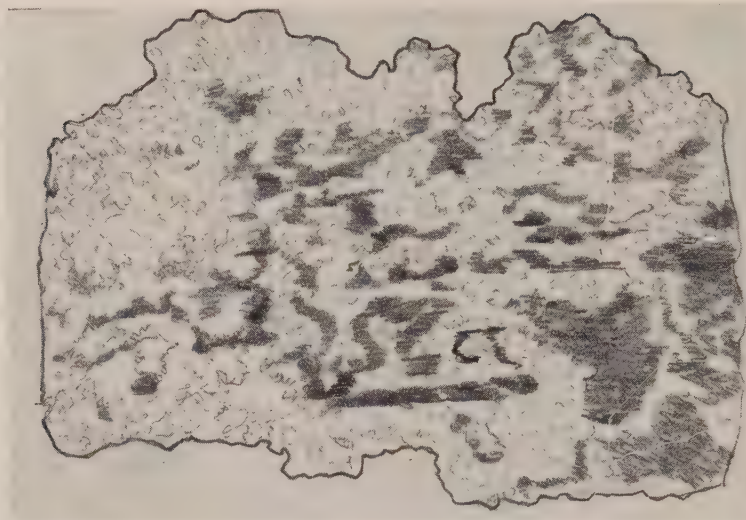


Fig. 19. Alteration of allemontite (dark) to arsenostibite (light). $\times 100$. M. Rosenberg del.

process having proceeded along two cleavage planes, one parallel to the elongation of the allemontite crystals, the other at right angles thereto. (Fig. 19.)

"Some difficulty was experienced in determining the density of arsenostibite, since the mineral is porous and often admixed with gangue. Several grains weighing between 10 mg and 20 mg were selected under the binocular microscope and their density determined by means of the Berman balance. The results ranged from 3.63 to 3.77, and the density of the mineral can be given as 3.7 ± 0.1 .

"When the powdered mineral is examined in immersion liquids under the microscope it appears as bright yellow transparent grains, which, however, are often turbid as a result of the presence of minute inclusions. The mineral is isotropic, and shows no trace of cleavage. The refractive index varies somewhat, ranging from 1.670 to 1.685; it was noticed that the higher the refractive index the stronger the yellow colour of the grains, and this phenomenon is presumably due to a variability in composition from grain to grain.

"X-ray powder photographs were made of the type material of arsenostibite, using Cu-K radiation and cameras of 114.6 mm diameter. These powder photographs agreed in all respects with those of stibiconite (Vitaliano and Mason, 1952); however, the lines in the arsenostibite photographs were somewhat diffuse, suggesting that the crystallites are very small. The length of side of the unit cube, a_0 , is 10.27 Å.

"The chemical analysis was made on material taken from some pieces of allemontite which were thickly coated with the arsenostibite. The carefully detached coating was, however, found to be full of minute quartz grains. It was not possible to free the analytical material from this quartz, which appears in the analysis as a relatively large percentage of insoluble material, without, however, disqualifying the results. The analysis was the work of Miss Thelma Berggren. Because of the small amount of material available, individual determinations were made on 0.1 g portions, and the results are therefore given to one decimal only.

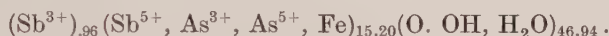
Table 11. Analysis of arsenostibite.

	1		2
Sb ₂ O ₅	45.4	Sb ⁵⁺	9.16
As ₂ O ₅	5.8	As ⁵⁺	1.66
Sb ₂ O ₃	4.3	Sb ³⁺	0.96
As ₂ O ₃	5.6	As ³⁺	1.84
Fe ₂ O ₃	6.2	Fe	2.54
Bi ₂ O ₃	0.3	O	34.96
CuO	0.0		
H ₂ O 105 ⁺	6.6	H ₂ O	11.98
H ₂ O 105 ⁻	8.1		
Insol.	18.1		
	100.4		

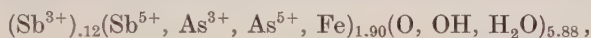
1: Weight percentages.

2: Atoms per unit cell.

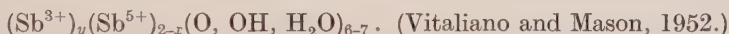
"When the analysis is recalculated into atoms per unit cell, by the procedure recommended by Hey (1939), the figures given in column 2 of Table 11 are obtained. If ions of similar size are grouped together, the analysed material can be formulated in the following way:



Dividing through by eight, we arrive at the following formula:



in good agreement with the general formula for stibiconite, which can be written



"Thus optical, X-ray, and chemical examination of the Varuträsk arsenostibite show that it is an arsenian variety of stibiconite, in which the proportion of antimony to arsenic is approximately 3:1."

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 VITALIANO, C. J. and MASON, B. (1952): *Am. Mineral.* 37, 982-999.

12. The triphylite-ferri-sicklerite-heterosite series

Triphylite

Summarized by BRIAN MASON

"Triphylite is the most common of the iron-manganese phosphate minerals found at Varuträsk. It is usually associated with cleavelandite and quartz. The fresh triphylite itself is pale gray in colour, but a thin coating of vivianite along the cleavage surfaces often gives it a deep sky-blue colour. Beginning oxidation to sicklerite causes it to acquire a wine-yellow to clove-brown colour. Individual crystals of triphylite have not been found, the mineral occurring in coarse-grained cleavable masses.

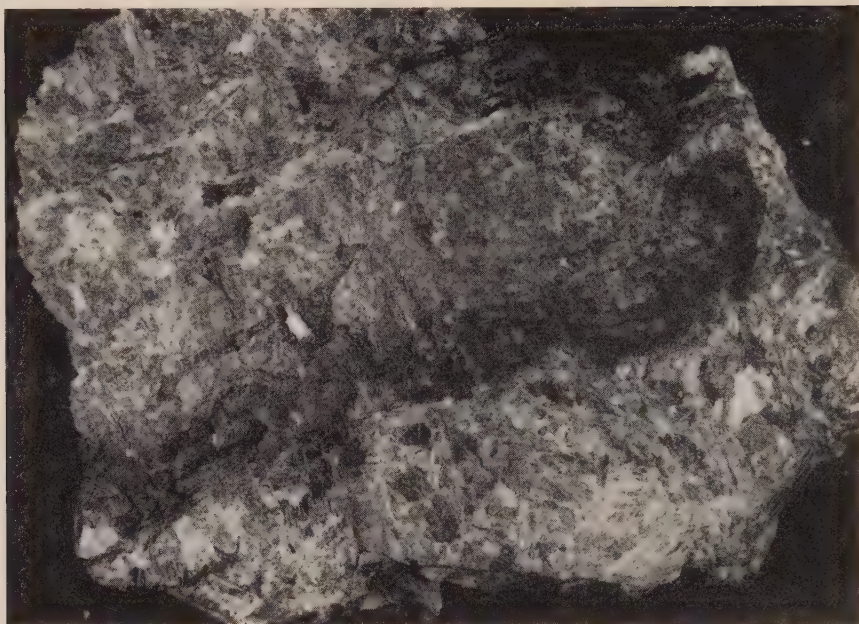


Fig. 20. Triphylite (upper part) with dark band of the oxidation products ferri-sicklerite and heterosite. Lower part of photo quartz-muscovite assemblage of the wall zone. Nat. size. Photo O. Mellis.

(Fig. 20.) The lustre is resinous, the fracture uneven to subconchoidal. The density of the analysed material (Anal. 1, Table 12) is 3.423.

"In thin sections the mineral is colourless and non-pleochroic. Small amounts of blue vivianite can often be seen, and beginning alteration to ferri-sicklerite appears as a brown colouration along the cleavages. The mineral is optically positive, with very strong dispersion, $r > v$; for the material of Anal. 1, Table 12, $2V = 26^\circ$, $\alpha = 1.686$, $\beta = 1.687$, $\gamma = 1.692$ (all measured in Na light). Quensel (1937) has observed twinning in one thin section of triphylite, and measurements by von Eckermann on the universal stage indicated that the symmetry of the mineral is lower than orthorhombic. However, an X-ray investigation by Björling and Westgren (1938) showed no deviation from orthorhombic symmetry.

"From X-ray powder photographs Björling and Westgren determined the lattice dimensions of the Varuträsk triphylite to be $a = 4.711 \text{ kX}$, $b = 10.374 \text{ kX}$, $c = 6.038 \text{ kX}$; from these figures the axial ratio is $0.454 : 1 : 0.583$. The powder photograph of triphylite is reproduced in Table 13 (d values in Å, not kX units).

"Three analyses have been made of the Varuträsk triphylite (Table 12, Nos. 1, 2, 3). The analytical work was carried out by Miss Thelma Berggren at the laboratories of the Boliden Mining Company. Analysis 1 is of perfectly fresh triphylite, analysis 2 is of wine-yellow triphylite surrounding the material of analysis 1 (Quensel 1937, fig. 1), while analysis 3 is of a yellow-brown triphylite found at an early stage of the investigation of the pegmatite. Optical examination of the analysed material shows that the content of Fe_2O_3 corresponds to the amount of ferri-sicklerite present. This is important in showing that the change from triphylite to ferri-sicklerite is

discontinuous rather than transitional, although both minerals have the same atomic structure, i.e. phases intermediate in composition between these two minerals are not formed. Analysis 1 corresponds well with the accepted formula of triphylite, Li(Fe,Mn)PO_4 ; the mineral contains 56 % of the LiFePO_4 component."

Ferri-sicklerite (*ferrian sicklerite*)

"Ferri-sicklerite was first described by Quensel (1937) on material from Varuträsk and from another pegmatite on the farm Lövlunden, about 3 km northeast of Varuträsk. The Varuträsk specimens show very clearly the relationship between ferri-sicklerite and triphylite, the ferri-sicklerite being pseudomorphous after triphylite and forming a shell which encloses a core of unaltered triphylite (Fig. 20). The change from triphylite to ferri-sicklerite evidently begins at the surface of the triphylite masses and gradually works inwards, until eventually the triphylite may be completely replaced by a mass of ferri-sicklerite which retains the form and the cleavage of the triphylite. When collected the surface of the ferri-sicklerite is dark brown to black; this is, however, a superficial coating and is easily removed by washing the specimen in warm dilute HCl, whereupon the true colour of the mineral, a light chestnut-brown, appears. Ferri-sicklerite has a brown streak; it has a dull, somewhat silky lustre. The density of the analysed material from Varuträsk is 3.391, that of the Lövlunden material 3.271 (determined by Miss Thelma Berggren).

"Under the microscope the ferri-sicklerite is pleochroic in shades of dark yellow to reddish brown. The refractive indices of the analysed material from Lövlunden are $\alpha = 1.785$, $\beta = 1.795$, $\gamma = 1.825$; pleochroism $X = \text{dark brown}$, $Z = \text{yellow brown}$; absorption $X > Y > Z$. The mineral is optically positive with an axial angle of about 60° .

"Björling and Westgren (1938) made an X-ray investigation of ferri-sicklerite from Varuträsk. They showed that the mineral has the same structure as triphylite; it is thus orthorhombic, and the cell dimensions are $a = 4.787 \text{ kX}$, $b = 10.086 \text{ kX}$, $c = 5.939 \text{ kX}$, giving an axial ratio of 0.475:1:0.588. The change from triphylite to ferri-sicklerite results in a slight decrease in the unit cell volume. The powder photograph of ferri-sicklerite is reproduced in Table 13 (d values in Å, not kX units).

"The analysis of ferri-sicklerite by Miss Berggren (Table 12, No. 4) shows that it is a lithium ferric manganous phosphate. The alteration of triphylite to ferri-sicklerite is evidently the oxidation of the iron from the ferrous to the ferric condition, with the removal of some of the lithium ions from the lattice to maintain electrical neutrality. The structure of ferri-sicklerite is the same as that of triphylite, but with ferric ions in the place of ferrous ions, and with the lithium lattice points partly vacant."

Heterosite

"Heterosite occurs at Varuträsk as nodules of variable size widely distributed through the pegmatite. One of the largest nodules measured over 40 cm in length and 20 cm in breadth. The nodules often contain cores of ferri-sicklerite and sometimes of triphylite also. Microscopic examination of such specimens shows clearly the paragenetic sequence from triphylite to ferri-sicklerite and then to heterosite; the individual minerals show, however, sharp boundaries one against the other, showing that the alteration is step-wise and not continuous.

"In the field heterosite always shows a dull black colour, which is due to thin superficial coating which is easily removed by scraping with a knife or by washing the specimen in warm dilute HCl. Such treatment reveals the magnificent purple of the heterosite itself. The streak is also purple. The lustre is dull and somewhat silky. The density of the analysed material (determined by Miss Thelma Berggren) is 3.409.

"Under the microscope the heterosite is practically opaque except in the thinnest grains. It shows intense pleochroism from yellow-gray to deep purple. The deep colouring makes optical determinations difficult. The mineral is fine-grained and it has not been possible to determine the sign of the axial angle.

"Björling and Westgren (1938) made an X-ray investigation of the heterosite from Varuträsk. They showed that the mineral has the same structure as triphylite; it is thus orthorhombic, and the cell dimensions are $a = 4.760 \text{ kX}$, $b = 9.680 \text{ kX}$, $c = 5.819 \text{ kX}$, giving an axial ratio of 0.492:1:0.601. The change from ferri-sicklerite to heterosite results in a further decrease in the volume of the unit cell. The powder photograph of heterosite is reproduced in Table 13 (d values in Å, not kX units).

"The analysis of heterosite from Varuträsk by Miss Thelma Berggren is given in Table 12 (No. 5). Microscopic examination of the analysed sample shows the presence of a small amount of admixed ferri-sicklerite, and it is believed that this admixture is responsible for the small amount of MnO and probably also for the Li_2O in the analysis. The analysis shows that the heterosite from Varuträsk is essentially a ferric-manganic phosphate, $(\text{Fe}, \text{Mn})\text{PO}_4$, containing 57 % of the ferric component and 43 % of the manganic component. When we compare this composition with that of the primary triphylite we can see that the oxidation from triphylite through ferri-sicklerite to heterosite has proceeded without change in the proportions of manganese and iron; the alteration has taken place in two steps, first the oxidation of the iron

Table 12. Analysis of triphylite, ferri-sicklerite, and heterosite from Varuträsk.

	1	2	3	4	5
Fe_2O_3	0.00	0.33	6.30	27.20	27.44
Mn_2O_3	—	—	—	0.00	20.42
Al_2O_3	0.00	0.00	0.00	0.00	—
FeO	24.33	22.85	18.21	0.59	—
MnO	18.95	18.15	20.16	19.13	3.60
CaO	1.82	1.84	1.36	1.36	1.70
MgO	trace	0.11	0.00	0.11	0.17
Li_2O	8.52	7.92	6.06	3.72	0.46
Na_2O	0.16	0.58	2.28	0.81	1.12
K_2O	0.00	0.00	0.00	0.00	0.00
P_2O_5	44.10	42.23	44.47	44.80	41.93
H_2O^+	0.31	0.36	0.64	0.51	1.65
H_2O^-	0.29	0.13	0.05	0.51	1.67
Insol.	1.24	5.80	0.20	1.66	0.30
Total	99.72	100.30	99.73	100.40	100.46
Sp. gr.	3.423	3.384	3.521	3.391	3.409

- 1: Fresh pale-gray triphylite.
- 2: Wine yellow triphylite.
- 3: Somewhat oxidized triphylite.
- 4: Ferri-sicklerite.
- 5: Heterosite.

All the analyses first published in G.F.F. 59, 1937, p. 81–88.

from the divalent to the trivalent state, then a similar oxidation of the manganese; at the same time lithium ions have been removed from the structure to maintain electrical neutrality. The final product of this process is heterosite, which retains the structure of the original triphylite in spite of the lattice positions originally occupied by lithium now being completely vacant."

Table 13. X-ray powder photographs of triphylite, ferri-sicklerite, and heterosite. Copper radiation, nickel filter.

Triphylite		Ferri-sicklerite		Heterosite	
I	d (Å)	I	d(Å)	I	d (Å)
30	5.22	40	5.04	30	6.28
80	4.29	80	4.35	10	5.51
30	3.95	20	3.83	20	4.85
80	3.51	80	3.48	60	4.29
10	3.33			30	3.87
80	3.03			80	3.48
40	2.79	80	2.96	20	3.06
90	2.54	20	2.75	40	2.93
20	2.47	90	2.50	60	2.73
10	2.39	30	2.45	10	2.51
20	2.29	10	2.33	30	2.43
10	2.15	10	2.25	10	2.32
10	1.86	10	2.17	10	2.16
10	1.81	10	1.87		
50	1.75	10	1.80		
10	1.67	20	1.75		
10	1.63	10	1.67		
10	1.59	30	1.61		
40	1.51	10	1.56		
10	1.35	10	1.48	20	1.46
10	1.31	10	1.35		
10	1.28				

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 MASON, B. (1941), Minerals of the Varuträsk Pegmatite XXIII. G.F.F. 63, p. 125, 135.

13. Lithiophilite

Summarized by BRIAN MASON

"Lithiophilite was described from Varuträsk by Quensel (1940) on the basis of material collected in 1939 and 1940. Lithiophilite is quite rare at Varuträsk and occurs in very much smaller amounts and in a rather different manner than its iron-rich analogue triphylite. The lithiophilite occurs in small nodules up to 3 cm in diameter (Fig. 21), surrounded by blue tourmaline and quartz in cleavelandite (Fig. 22). The mineral is light yellow-green to olive green in colour, and has a characteristic resinous lustre. In some specimens the lithiophilite shows rims of oxidation, evidently the normal sequence of sicklerite and purpurite (Fig. 23).



Fig. 21. Lithiophilite in cleavelandite. Excavation G₂. $\times 2$. Photo O. Mellis.

"In thin sections under the microscope the lithiophilite is colourless. The characteristic pinacoidal cleavages parallel to (001) and (010) are well developed. The mineral is positive, the axial angle $2V_{Na} = 64^\circ$ (not $2E_{Na}$ as in the original description). Dispersion marked, $r > v$. The indices of refraction are $\alpha = 1.660$, $\gamma = 1.678$. The X-ray powder photograph is identical with that of triphylite.

"The analysis of lithiophilite was made by Miss Berggren on 0.4 g of material from a single specimen found in 1939. Because of the small amount of material used for the analysis the results are reported to one place of decimals only. The lithiophilite could not be completely separated from the quartz and cleavelandite with which it is intimately intergrown, and these materials account for the 10.3% insolubles reported in the analysis. The analysis is in Table 14.

"This analysis resembles closely one of a lithiophilite from Norway, Maine (Penfield, 1883). It is noteworthy, however, for the unusually high content of Na_2O and CaO , which are usually absent or present in little more than trace amounts in lithiophilite."

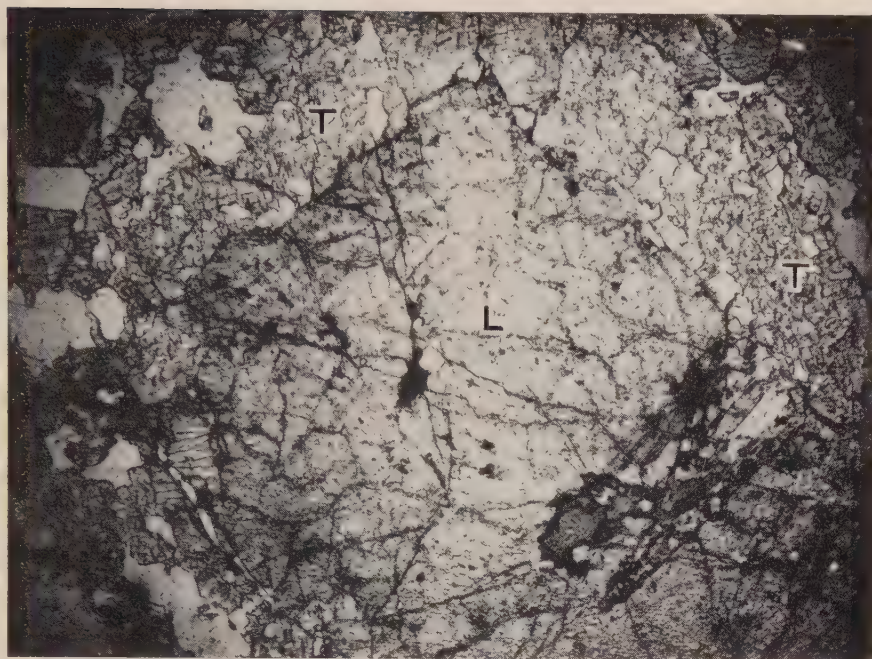


Fig. 22. Nodule of lithiophilite (L) showing rim of indicolite and quartz in poikilitic intergrowth (T). $\times 25$. Photo W. Uytenbogaardt.

Table 14.

Li ₂ O	6.7
Na ₂ O	2.4
K ₂ O	0.5
FeO	6.4
MnO	29.9
CaO	3.0
P ₂ O ₅	40.5
H ₂ O	0.3
Insol.	10.3
	<hr/>
	100.0

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 MASON, B. (1941), Minerals of the Varuträsk Pegmatite XXIII. G.F.F. 63, p. 125, 135.

14. The varulite-alluaudite-purpurite series

Varulite

Summarized by BRIAN MASON

"The original material of the new mineral varulite was collected by Miss Thelma Berggren at Varuträsk in 1936 and described by Quensel in 1937. In 1940 he published two new analysis of this mineral and discussed its chemical formula and its relation



Fig. 23. Lithiophilite, partially oxidised to purpurite (black). $\times 80$. Photo W. Uytendogaardt.

to the chemically similar mineral arrojadite. In 1941 Mason published a fourth analysis of varulite from Varuträsk and further discussed its formula and mineral relationships, suggesting that it might be considered a natrophilite in which some of the sodium was replaced by calcium. In 1950 Lindberg discussed the relationship between varulite and arrojadite and showed by X-ray investigation of type material that these two minerals are structurally distinct, although they are rather similar in chemical composition. She also showed that natrophilite is structurally distinct from both arrojadite and varulite, and that Mason's suggestion of a close relationship between the three species was thus untenable.

"Varulite occurs at Varuträsk as nodules seldom more than two centimeters in diameter, generally embedded in a matrix of quartz and muscovite (Fig. 24). The nodules are usually rounded but rare specimens show an approximation to what may be crystal outlines. In hand specimen the colour of the mineral is olive-green and the lustre is dull. The streak is gray-green. The density of the original material (Anal. 3, Table 15) is 3.58.

"Quensel (1937) gives the following description of the optical properties of varulite: 'Under the microscope the mineral is colourless. Two well-marked cleavages at

right-angles probably are the same as in the triphylite series, (001) and (010). The axial plane is, however, vertical to the best cleavage. If this also here is (001), the plane would be parallel (010). Refractive indices are $\alpha = 1.720$, $\gamma = 1.732$. The axial angle is distinctly larger than in triphylite, $2V_{Na}$ is 70° . Dispersion distinct $r > v$, but not nearly as strong as in triphylite. Most grains are intimately interwoven with a yellowish green phosphate, without doubt alluaudite, representing beginning alteration to that mineral.'

'Microscopic examination of material of the analysed varulites from Varuträsk shows that they consist of an intimate intermixture of two minerals, one colourless and the other green. The green mineral replaces the colourless mineral and is evidently an alteration product of it. The optical properties reported by Quensel refer to the colourless mineral, with the exception of the refractive indices, which are those of the green mineral. Hence the statement by Lindberg (1950, p. 71) referring to the varulite from Varuträsk: 'No mineral, colourless under the microscope, with indices $\alpha = 1.720$ and $\gamma = 1.732$ was noted.'

'A few grains of the colourless mineral were isolated from an analysed varulite (Anal. 1, Table 15) and the indices were found to be $\alpha = 1.660$, $\gamma = 1.673$. An X-ray powder photograph of this material proved to be identical with that of lithiophilite and the refractive indices confirm this identification.

'One thin section shows lithiophilite in contact with yellow-brown alluaudite, the latter mineral containing small grains of green varulite. The lithiophilite also encloses many small grains and streaks of varulite. The oxidation has here been sufficiently intense to convert the lithiophilite into alluaudite with only an insignificant amount of the varulite surviving. (Fig. 25.)

'From Quensel's original description it appears that he intended the name varulite to apply to the colourless mineral, rather than the green mineral. However, the name has also been used for the green mineral and the data in the literature for the refractive indices and the pleochroism refer to this substance. The colourless mineral now proves to be lithiophilite. I therefore propose that the name varulite be applied to the green mineral; it is different in structure, in chemical composition, and in physical properties from lithiophilite; it is structurally similar to alluaudite (judging from X-ray powder photographs), but is distinct in its green colour (alluaudite is yellow-brown), in its physical properties, and in the presence of most of the iron in the bivalent state. At Varuträsk varulite is an alteration product of lithiophilite, just as the structurally similar hühnerkobelite (Lindberg, 1950) is an alteration product of triphylite at Hühnerkobel (Mason, 1942) and at Norrö (Eriksson, 1946).

'In Table 16 X-ray powder spacing data are given for the bulk material of Anal. 1, Table 15, which consists of varulite (as here defined) with a small admixture of lithiophilite, the lithiophilite being apparently insufficient in amount to give its characteristic lines on the powder photograph.

'In all, four analyses have been made on varulite from Varuträsk, and they are given in Table 15. They are all quite similar, the most noticeable difference being in the percentages of FeO and Fe₂O₃. The sum of these two oxides is, however, relatively constant. Examination of the analysed material shows that it contains in each case predominantly the green mineral, with smaller and variable amounts of lithiophilite. The lithium content shown by the analyses is evidently derived from this admixed lithiophilite.

'Both Quensel (1937, 1940) and Mason (1941) have attempted to derive from the analyses a formula for 'primary, unoxidised varulite'. Quensel originally suggested

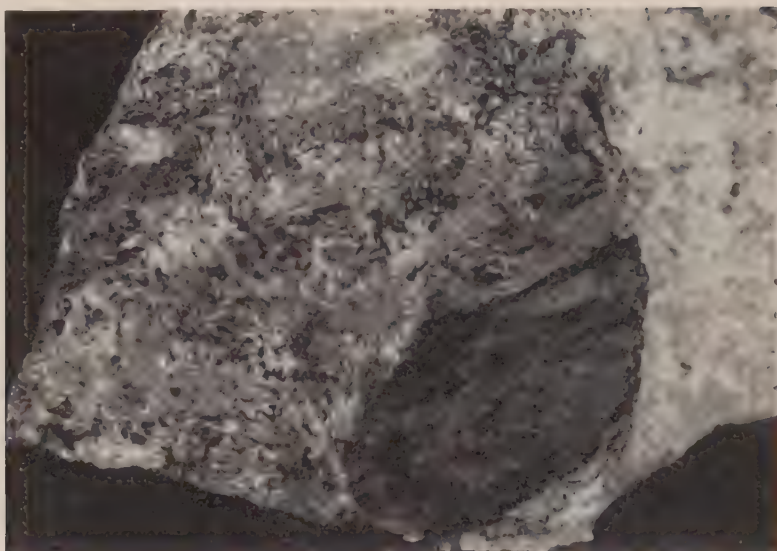


Fig. 24. Nodule of varulite. Central part unaltered varulite. The darker rim is the oxidation product purpurite. Nat. size. Photo O. Brotzen.



Fig. 25. Lithiophilite, partially altered to alluaudite (black). Disseminated shadowed areas in lithiophilite are remnants of varulite, representing the intermediate stage of oxidation. $\times 80$. Photo W. Uytendogaardt.

$\text{Na}_2\text{O} \cdot 5\text{RO} \cdot 2\text{P}_2\text{O}_5$, R representing the bivalent metals; Mason preferred to group the sodium and calcium together, and arrived at the formula $(\text{Na}_2, \text{Ca})0.2(\text{Mn}, \text{Fe})\text{O} \cdot \text{P}_2\text{O}_5$. This latter formula suggests a close relationship between varulite and natrophilite, a suggestion which is not borne out by the later work of Lindberg. A reexamination of the analyses shows that they can also be interpreted in terms of a formula $(\text{Ca}, \text{Na})_2(\text{Mn}, \text{Fe})_3(\text{PO}_4)_3$. However, until more is known of the crystal structure of varulite the above considerations of its chemical formula are largely speculative."

Table 15. Analyses of varulite and alluaudite from Varuträsk.

	1	2	3	4	5	6
Fe_2O_3	9.29	5.32	8.35	7.66	19.83	16.44
FeO	9.73	11.03	7.52	9.50	—	1.98
MnO	25.43	25.31	25.30	24.76	28.17	26.24
CaO	2.51	2.30	4.86	2.75	—	2.16
MgO	trace	0.13	0.00	0.16	1.19	0.00
Li_2O	0.46	1.65	0.88	0.53	—	0.28
Na_2O	8.10	8.08	7.12	9.02	7.20	6.94
K_2O	trace	0.12	0.12	0.07	—	0.13
P_2O_5	43.40	44.93	42.80	44.14	40.70	42.76
F	0.00	0.00	0.06	0.00	0.80	0.00
H_2O^+	1.16	0.64	0.75	0.60	0.66	1.60
H_2O^-	0.07	0.18	0.14	0.15	0.24	0.88
Insol.	0.18	0.28	1.80	0.24	1.24	0.54
Less O for F . . .				0.19		
Total:	100.33	99.97	100.06	99.58	100.03	99.95
Sp. gr.			3.581		3.576	

Cl = tr. $\text{Al}_2\text{O}_3 = 0.36$ Cl = tr.

1: Varulite.	Minerals of the Varuträsk Pegmatite	XXIII.	G.F.F. 63, 130.
2: "	" " "	"	XVII. G.F.F. 62, 298.
3: "	" " "	"	I. G.F.F. 59, 94.
4: "	" " "	"	XVII. G.F.F. 62, 298.
5: Alluaudite.	" " "	"	I. G.F.F. 59, 92.
6: "	" " "	"	XVIII. G.F.F. 62, 300.

Table 16. X-ray powder photographs of varulite and alluaudite. Copper radiation, nickel filter.

Varulite		Alluaudite	
I	d (Å)	I	d (Å)
70	6.37	60	6.33
10	5.50	30	5.44
		10	4.17
70	3.53	30	3.48
40	3.12	40	3.07
20	2.92		
		20	2.85
100	2.75	100	2.72
70	2.56	50	2.51
10	2.13	40	2.09
20	1.93		
20	1.76	10	1.75
20	1.68		
		10	1.58

Alluaudite

"The mineral alluaudite was described over a century ago from a pegmatite at Chanteloube in Central France, but this remained the single occurrence until Quensel in 1937 identified it in material from Varuträsk. It occurs as a thin coating on varulite. On the weathered surface the alluaudite may be dull yellow to practically black, but washing in warm dilute hydrochloric acid brings out the characteristic canary yellow colour of the mineral. The streak is yellow-brown. The density of the Varuträsk alluaudite (Anal. 5, Table 15) is 3.576.

"Under the microscope the Varuträsk alluaudite appears as exceedingly fine-grained aggregates with a yellow to yellow-brown colour. It has not been possible to determine the sign or axial angle. The refractive indices are $\alpha = 1.760$, $\gamma = 1.775$.

"The identity of the Varuträsk mineral with the original alluaudite from Chanteloube has been confirmed by comparing X-ray powder photographs, which are identical. The X-ray spacing data for the mineral are given in Table 16.

"Two analyses have been made of the alluaudite from Varuträsk, and they are reproduced in Table 15. They are quite similar and show that alluaudite is essentially a sodium ferric manganous phosphate. Alluaudite is related to varulite in the same way as ferri-sicklerite is related to triphylite, i.e. the alteration of varulite to alluaudite involved the oxidation of the iron from the ferrous to the ferric state, with the concomitant removal of some sodium ions from the lattice to maintain electrical neutrality."

Purpurite

"Purpurite, the manganese analogue of heterosite, occurs at Varuträsk in association with alluaudite, from which it has evidently been formed by oxidation. When collected in the field, specimens have a dark brown or black coating which can be removed by scraping or by washing in warm dilute hydrochloric acid; after such treatment the brilliant purple colour of the mineral is visible, and the purpurite is generally seen to form a peripheral shell around nodules of alluaudite.

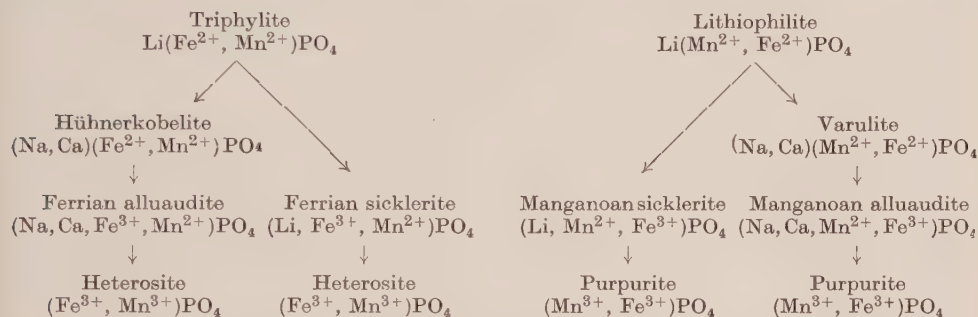
"Under the microscope the purpurite is practically opaque except in the thinnest grains. Pleochroism is very strong in shades of wine-red and magenta. The mineral is fine-grained and it has not been possible to determine the sign or the axial angle.

"No chemical analyses have been made of the purpurite from Varuträsk. However, its paragenetic relationship to the manganese-rich alluaudite indicates that it too should be a manganese-rich phosphate and therefore specifically classed with purpurite.

"In the original description of purpurite from Varuträsk, Quensel (1937) applied the name Na-purpurite to it to indicate that it had been formed from the sodium-bearing phosphates varulite and alluaudite, and not as previously described purpurites from the lithium-bearing lithiophilite. Björling and Westgren (1938) investigated the Varuträsk purpurite by means of X-ray powder photographs and wrote as follows: '... Their X-ray powder photographs resemble neither those of varulite or Mn-alluaudite, nor those of heterosite. They contain a very large number of lines, proving that the structure of Na-purpurite is exceedingly complicated.' However, Mason (1941) was able to show that the material X-rayed by Björling and Westgren as Na-purpurite was actually a mixture of alluaudite and purpurite, and the complicated powder pattern was due to the presence of these two phases. Pure purpurite derived from alluaudite gave an X-ray pattern differing in no way from that of purpurite derived from lithiophilite."

According to Mason's renewed investigation, varulite must now be taken as in itself an alteration product of lithiophilite. It therefore is evident that both lithiophilite and triphylite succumb to alterations along two different lines, both terminating in the related end products purpurite and heterosite. On the one hand the intermediate stage is represented by the minerals of the sicklerite group. On the other hand two intermediate mineral stages are present. At Varuträsk they are varulite and alluaudite as intermediate phases in the series lithiophilite–varulite–alluaudite–purpurite. In a pegmatite on the small island Norrö in the archipelago of Stockholm hühnerkobelite (in the original description named arrojadite, T. Eriksson 1946) and alluaudite represent similar stages intermediate in the series triphylite–hühnerkobelite–alluaudite–heterosite.

The probable sequence of these parallel alterations is displayed in the scheme below.



The cause, leading to the different trends of alteration, as seen in the plan above, must be that in the one case, represented by the series triphylite–ferrian sicklerite and lithiophilite–manganoan sicklerite, no exchange of alkali components has taken place. In the other case, represented by the series triphylite–hühnerkobelite–ferrian alluaudite and lithiophilite–varulite–manganoan alluaudite, lithium has been substituted by sodium and calcium. The final products of both series are, however, identical, in the triphylite series ending in heterosite, in the lithiophilite series in purpurite.

REFERENCES

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 ERIKSSON, L. (1946), Arkiv för Kemi etc. 23 A, No. 8.
 LINDBERG, M. L. (1950), Am. Mineral. 35, 59–76.
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15. Triplite

Summarized by BRIAN MASON

“Triplite is the rarest of the iron-manganese phosphates found at Varuträsk. A single specimen of this mineral was collected on the dumps in 1939 and was described by Mason in 1941.¹ Hence there is little to add to the original description.

¹ Since then a second, larger sample of triplite has been found. It was, however, much decomposed and crusted with vivianite.

"The single specimen consists of narrow veins of triplite, up to five millimeters broad, in a matrix of quartz and mica. The whole specimen is covered with a thin blue coating of vivianite, evidently a secondary product formed from the triplite. The colour of the triplite itself is chestnut brown: the lustre vitreous to resinous; the mineral is massive and shows no sign of crystal outlines. The density is 3.84. The optical properties of the analysed material are $\alpha = 1.673$, $\beta = 1.681$, $\gamma = 1.691$, (+), $2V = 76^\circ$.

"The chemical analysis by Miss Berggren of the triplite is given in Table 17. The analysed sample was contaminated with some quartz and mica, but this contamination had no effect on the main analysis, as when the sample was warmed for some minutes with dilute nitric acid all the triplite dissolved, leaving the quartz and mica as insolubles (6.22 %). The determination of the alkalis by the Lawrence Smith method, and the determination of fluorine and water, were made on portions of the main sample, and thus some of the potassium, and a little of the fluorine and water, may have been derived from the small amount of mica present as impurity.

Table 17. Analysis of triplite.

	1	2
Fe ₂ O ₃	0.78	
FeO	19.96	0.2778
MnO	32.07	0.4522
CaO	4.68	0.0834
MgO	trace	
Li ₂ O	0.10	
Na ₂ O	0.39	
K ₂ O	0.33	
P ₂ O ₅	30.63	0.2156
F	6.72	0.3537
H ₂ O ⁺	0.48	0.0532 (OH)
H ₂ O ⁻	0.27	
Insol.	6.22	
	102.63	
Less O for F	2.83	
	99.80	

"When the analysis is calculated in molecular proportions (2) the RO ratio (R = Mn, Fe, Ca):P₂O₅:(F, OH) is 4.00:1.06:2.00, corresponding well with the accepted formula of triplite, R₂FPO₄. R is approximately Mn_{.56} Fe_{.34} Ca_{.10}. 13 % of the fluorine is replaced by hydroxyl.

"X-ray powder photographs of the triplite from Varuträsk agree with the measured powder photograph published by Lindberg (1950)."

REFERENCES

- LINDBERG, M. L. (1950), *Am. Mineral* 35, 72.
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16. Amblygonite (Montebrasite)

The analysed samples of amblygonite from Varuträsk all refer to the sub-species montebrasite in somewhat varying modifications.

An older generation, pertaining to the pegmatitic stage, occurs in large anhedral

masses or in huge crystals up to 15 dm in diameter. Crystal faces are generally only indistinctly developed.

A second generation of the same mineral, pertaining to a later stage of mineralisation, on the other hand occurs in much smaller individuals, seldom more than 5 cm in length.

Both modifications are milky white in colour. The former type has a dull greasy lustre whereas the latter type has a vitreous lustre. A single specimen was found to have a light salmon colour.

Optical determinations show no divergences between these different types. Polysynthetic twinning is nearly always observable, often in a very fine-laminary development. The indices of refraction for the second modification (analysis 2) are $\alpha = 1.5679$, $\beta = 1.6093$, $\gamma = 1.6199$. $2V_{Na} = 81^\circ 30'$.

S. Tengnér undertook to measure some crystals and could identify the forms (001), (100), (010), (110), ($\bar{1}10$), ($\bar{1}\bar{1}0$), ($\bar{1}20$), ($\bar{1}\bar{2}0$), (0 $\bar{1}1$), (0 $\bar{2}1$), and ($\bar{1}01$). The forms *f* (0 $\bar{1}1$) and *b* (010) have not previously been recorded on amblygonite. As the form (010) is known on fremontite, a partial analysis was executed on material from the measured crystal to verify its composition. The result gave Li₂O 9.68 and Na₂O 0.30. The alkali components correspond with those of montebrasite in analysis 1 of Table 18.

Three analyses of the three different modifications of the mineral are given in Table 18. The only difference of any importance in chemical composition is the varying percentage of F and H₂O in analyses 1 and 2 and a somewhat higher content of Na₂O in analysis 3.

Table 18.

	1	2	3
Al ₂ O ₃	34.91	—	34.39
Fe ₂ O ₃	0.10	—	0.04
FeO	0.03	—	—
MnO	0.00	—	—
CaO	0.00	—	—
MgO	0.00	—	—
Li ₂ O	9.98	9.41	9.14
Na ₂ O	0.38	0.95	2.17
K ₂ O	0.00	—	—
P ₂ O ₅	47.99	—	47.79
F	2.10	4.42	1.80
H ₂ O ⁺	5.22	3.71	5.20
H ₂ O ⁻	0.04	—	0.09
insoluble	0.00	—	0.10
	100.75		100.72
—O for F	0.88		0.76
	99.87		99.96
Sp. gr.	3.025	3.027	3.002

- 1: Montebrasite, large crystal of first generation. Th. Berggren anal.
 2: Montebrasite, small crystal of second generation (partial analysis).
 3: Montebrasite, salmon coloured type. Analyses 2 and 3 by R. Blix.

Signs of superficial decomposition are not present. A well-defined zone of reaction is on the other hand frequently to be observed in the form of a narrow rim, surrounding nodules of fresh montebrasite encompassed in cleavelandite. The reaction rim, only

2–3 mm in breadth, consists of a dense, pale pink material, sometimes bordered by an outer rim of lepidolite. (See Fig. 65, p. 112.) Under the microscope the inner rim is found to consist of minute flakes of mica intermixed with a kaolin mineral. Landes has described similar conditions from a Main pegmatite (Landes 1925).

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 QUENSEL, P. (1937), *Minerals of the Varuträsk Pegmatite VIII*. G.F.F. 59, p. 455, with crystallographic data by S. TENGNÉR.
 TENGNÉR, S. (1940), *Minerals of the Varuträsk Pegmatite, XVIII*. G.F.F. 62, p. 352.

17. Manganapatite

Manganapatite is of frequent occurrence at Varuträsk. It is generally associated with dark quartz and green tourmaline. In a massive lepidolitic rock inclusions of manganapatite are often abundant.

The mineral is mostly developed as short, thick prismatic crystals up to 3 cm in diameter, roughly bounded but without defined faces. The colour is dark bluish green. The indices of refraction, determined on refractometer in sodium light are: $\alpha = 1.6459$ $\gamma = 1.6411$.

An analysis is given in Table 19.

Table 19.

	1		1
Al ₂ O ₃	0.00	K ₂ O	0.00
Fe ₂ O ₃	—	P ₂ O ₅	41.50
FeO	0.26	F	3.41
MnO	5.32	H ₂ O ⁺	0.25
CaO	50.31	H ₂ O ⁻	0.03
MgO	0.04	Insol.	0.35
Li ₂ O	0.00		101.47
Na ₂ O	0.00	— O for F	1.56
Sp. gr.	3.22		99.91

1: Manganapatite Varuträsk, R. Blix anal.

REFERENCE

- QUENSEL, P. (1937), *Minerals of the Varuträsk Pegmatite*. G.F.F. 59, p. 157.

18. Hydroxylapatite (*manganooan*)

Summarized by BRIAN MASON

"This mineral was originally described by Mason in 1941 as manganhydroxyapatite. In the seventh edition of Dana's System of Mineralogy it is referred to as a manganooan variety of hydroxylapatite and in the interest of uniformity in nomenclature this designation is accepted here.

"The manganooan hydroxylapatite occurs as small veins and irregular segregations in specimens of varulite. Its white colour causes it to show up very clearly against the dark green varulite. It is a rare mineral, present only in some few varulite specimens, and varulite itself is not common. Study of the specimens suggest that the hydroxylapatite is later than the varulite and replaces it.

"The hydroxylapatite is very fine-grained and shows no trace of crystal form or cleavage. The colour is generally pure white; occasional specimens show a bluish tinge. The mineral is not transparent even in thin splinters; in thin sections it is translucent. The lustre is dull and greasy. The hardness is 5 and the density 3.19.

"Thin sections under the microscope show that hydroxylapatite is uni-axial and negative. The refractive indices determined in sodium light by the immersion method are $\varepsilon = 1.657$, $\omega = 1.661$.

"Considerable time was spent in obtaining a pure sample of the hydroxylapatite for chemical analysis. The material was carefully handpicked under binocular microscope. A sample of 0.3 g was obtained, and this sample was analysed by Miss Berggren. The analysis give the following results:

Table 20.

	1	2
CaO	44.14	45.78
MnO	7.50	7.58
FeO	3.07	3.49
MgO	0.00	
SrO	0.18	
Li ₂ O	trace	
Na ₂ O	0.58	
P ₂ O ₅	40.26	41.40
F	0.00	
CO ₂	0.00	
H ₂ O ⁺	2.56	1.75
H ₂ O ⁻	0.31	
insoluble	0.49	
	99.09	100.00

1: Hydroxylapatite (manganoan). Varuträsk.

2: Composition calculated for $\text{Ca}_{8.4}\text{Mn}_{1.1}\text{Fe}_{0.5}\text{P}_6\text{O}_{24}(\text{OH})_2$.

"On account of the small sample, the second decimal figure in the percentages must be considered uncertain. Chlorine and sulphur were sought for in a separate sample, but were not found. The content of lithium, sodium, and strontium was determined spectrographically.

"When the analysis is recalculated in terms of atoms the formula is $\text{Ca}_{8.4}\text{Mn}_{1.1}\text{Fe}_{0.5}\text{P}_6\text{O}_{24}(\text{OH})$. Both the manganese and the iron content are high in comparison with other analyses of apatite.

"X-ray powder photographs were taken of the hydroxylapatite, using chromium radiation. Over 20 reflections were measured and could be indexed in terms of the apatite structure. The cell dimensions thus obtained were $a_0 = 9.54 \text{ kX}$, $c_0 = 6.73 \text{ kX}$, $a_0:c_0 = 1:0.705 \times \text{cell volume}$ ($a^2c \sin 60^\circ \times 10^{24} \text{ cm}^3 = 530$). The molecular weight calculated from the analysis is 1029. Using this value, the theoretical density for the lattice dimensions given above is 3.20 The measured density is 3.19.

"The occurrence of hydroxylapatite at Varuträsk is interesting from a geochemical standpoint. Its presence suggests that at the time of its deposition there was a practical absence of fluorine and chlorine in the mineral deposition fluids. It appears that at no time during the deposition of the minerals of the pegmatite there was an excess of fluorine present. The manganapatite is often very deficient in fluorine, giving rise to an oxyapatite (voelckerite). The amblygonite is also of the hydroxyl or montebasite type, never containing as much as 40 % of the $\text{Li}(\text{Al}, \text{F})\text{PO}_4$ component

and generally containing much less. The occurrence of a pure hydroxylapatite is noteworthy therefore as providing further evidence for the apparent deficiency of fluorine in the medium of deposition."

REFERENCE

MASON, B. (1941), Minerals of the Varuträsk Pegmatite XXVI. G.F.F. 63, p. 279.

19. Manganvoelckerite (*manganoan oxiapatite*)

Besides manganapatite in development as advanced above, a dissimilar type is occasionally to be found. Attention was drawn to this modification on account of the fact that on exposed surfaces it was always found to occupy well-defined cavities, evidently due to weathering of the mineral (Fig. 26). As manganapatite in normal development never shows any tendency to weather in this way, a mineral of some difference in composition was assumed existent.

Another difference is further noticeable in so far as the mineral is of a lighter blue colour than the usual form of manganapatite. In some specimens one can observe a kernel of deeper blue manganapatite in normal development surrounded by a zone of the lighter blue mineral. This suggests that during the mineral deposition a change in the composition of the solutions has taken place. As (OH) is present in about the same amount as in the analysis of the manganapatite, the difference must be ascribed to the content of fluorine, which according to the analyses has decreased from 3.41 % in manganapatite to 1.43 in the new modification.

The analysis (Table 21) indicates that at Varuträsk an oxyapatite of this type is present. The name manganvoelckerite has been taken as applicable in this instance.

The refractive indices are not very different from those of manganapatite. Comparison is given below:

Manganapatite	$\omega = 1.6459$	$\varepsilon = 1.6411$
Manganvoelckerite.	$\omega = 1.6492$	$\varepsilon = 1.6365$

In the new edition of Dana's System of Mineralogy (1951) the existence of oxyapatites, including voelckerite, is doubted and their presumed occurrence is ascribed to incomplete or defective analyses. Professor F. E. Wickman of the Mineralogical Dept. of the Natural History Museum in Stockholm kindly undertook to investigate how far this can be considered as justified. The results have kindly been put at my disposal and follow below:

"Professor Quensel has asked me to give my opinion on the voelckerite problem. As is well known, the name voelckerite was proposed by A. F. Rogers (1912) to designate a mineral with the hypothetical composition $3\text{Ca}_3(\text{PO}_4)_2 \cdot \text{CaO}$ as well as apatites where this component predominates. The pure mineral has never been observed or synthesized. In 1937 Quensel described a mineral from Varuträsk for which he proposed the name manganvoelckerite. In 1941 B. Mason showed that a mineral described as mangualdite was identical with Quensel's manganvoelckerite.

"Before the structural aspect of the problem is discussed I should like to touch upon another aspect of it. Three members of the apatite group have been found at Varuträsk and all have distinctly different colours.

	Colour	MnO	FeO	F	Cl	H ₂ O
Manganapatite	dark blue	5.32	0.26	3.41	—	0.25
Manganvoelckerite . .	light blue	4.31	0.32	1.43	0.02	0.17
Mangan-hydroxylapatite	white	7.50	3.07	0.00	—	2.56



Fig. 26. Manganvoelckerite, deeply hollowed by weathering, thereby contrasting to normal manganapatite. $\times 1.5$. Photo O. Mellis.

"The hydroxylapatite is white in spite of the fact that it contains about twice as much manganese and iron as the other two minerals manganapatite and manganvoelckerite, which have blue colours. If the colours are to be related to the chemical composition, a glance at the table shows that it should be related to the fluorine content. For physical reasons this conclusion seems not likely. It might be suspected, however, that the oxidation states of iron and manganese have not been determined correctly and that this might be the solution to the problem. Dr. R. Blix kindly tested this hypothesis. A specially designed apparatus was used to dissolve the mineral in HCl without any traces of atmospheric oxygen being present. Any chlorine gas that might be found by this process was brought into reaction with a solution of KI by a special distillation operation. The iodine formed was determined by the thiosulphate method. Blank runs and determinations on chemically well-defined compounds proved the reliability of the method which will be described by Blix in another paper. In the present case it was proved that within the limits of error, i.e. less than 0.01 %, no manganese or iron is present in a higher valency state than two in manganvoelckerite or manganapatite. It must therefore be concluded that the reason for the remarkable colours of the apatite minerals in Varuträsk is unknown.

"As already mentioned it has been suggested that all analyses indicating the existence of voelckerites should be regarded as unreliable. Personally, I have no

opinion about voelckerite analyses in general but I think that the two analyses on manganvoelckerite performed by Miss Berggren for Quensel and Mason are reliable for the following reasons:

"1. Apatite and voelckerite on a weathered rock surface behave quite differently. The apatite is intact but the voelckerite dissolves easily forming cavities in the rock surface (see Fig. 26). Actually this is how it was discovered.

"2. The analysed material was carefully controlled under the microscope, this constituting a guarantee of its purity.

"3. Miss Berggren, an experienced mineral analyst, has also performed a number of analyses on other members of the apatite group and in all cases—except the two voelckerites—the resulting analyses have been in excellent agreement with the ordinary apatite formula.

"Therefore in my opinion we have to accept the existence of members of the apatite group which correspond to the voelckerite definition. Of course this does not mean that Rogers' original end member necessarily should exist.

"If we accept the existence of voelckerite the next task is to describe the compound in terms of the apatite structure. D. McConnell (1952) touches on the problem but he concludes that it still remains to be solved.

"The well-known structure of apatite (cf. Strukturbericht Vol. 2) has the following atomic parameters.

Space group: $P6_3/m$. Unit cell dimensions: $a = 9.37 \text{ \AA}$; $c = 6.88 \text{ \AA}$.

Atomic positions:

- 4 CaI in $(f) \pm (\frac{1}{3} \frac{2}{3}, z; \frac{1}{3} \frac{2}{3}, \frac{1}{2} - z); z = 0$
 6 CaII in $(h) \pm (x \ y \ \frac{1}{4}; \bar{y}, x - y, \frac{1}{4}; y - x, \bar{x}, \frac{1}{4}); x = \frac{1}{4}; y = 0$
 6 P in $(h) \ x = \frac{5}{12}; y = \frac{13}{36}$
 6 OI in $(h) \ x = \frac{1}{3}; y = \frac{1}{2}$
 6 OII in $(h) \ x = \frac{3}{8}; y = \frac{7}{15}$
 12 OIII in $(i) \pm (x \ y \ z; \bar{y} \ x - y, z; y - x, \bar{x}, z; x, y, \frac{1}{2} - z; \bar{y}, x - y, \frac{1}{2} - z; y - x, \bar{x}, \frac{1}{2} - z)$
 $x = \frac{1}{3}; y = \frac{1}{4}; z = \frac{1}{16}$
 2 F in $(a) \pm (00 \ \frac{1}{4})$
 2 Cl in $(b) \ 000; 00 \ \frac{1}{2}$.

"A remarkable property of this structure is the difference between fluorapatite and chlorapatite. The fluorine and chlorine atoms occupy in fact different positions in the channels at the origin which run through the structure parallel with the c -axis. The fluorine atom (and the hydroxyl group) is comparatively small and so it occurs in the same level as the surrounding CaII atoms to which it is bonded. The chlorine atom on the other hand is too large to occupy these positions, and therefore it occurs at a distance half-way between the CaII atoms where the 'channel' is wider. As a consequence it is bonded to CaII atoms in two different levels.

"There is no indication that the voelckerite structure should be essentially different from the apatite structure. At least this must be true for the main framework of the structure which is built up of the phosphate groups and the calcium-manganese atoms. Our interest might therefore be concentrated on the channels in the structure. It seems plausible to assume that some of these channels are occupied by fluorine and hydroxyl in their ordinary position, i.e. $z = \frac{1}{4}$ and $z = \frac{3}{4}$. In other channels chlorine atoms occupy their ordinary position, i.e. $z = 0$ and $z = \frac{1}{2}$.

"Before the position of the oxygen atoms is discussed, the chemical analyses of

manganvoelckerite must be recalculated (Tables 21 & 22). It is assumed that Ca, Mn, Fe^{2+} , Na and K occupy the calcium positions of the apatite structure and that the phosphorus atoms exclusively occupy the P positions. This means that Li, Mg, Al and Fe^{3+} are to be regarded as impurities. In the tables it has been assumed that the number of large cations is exactly 10.00 or alternatively the number of phosphorus atoms is exactly 6.00. In both cases there is a surplus of oxygen atoms and a deficit of univalent anions. If we calculate the number of charges we find

	Varuträsk	Mangualde
Large cations = 10.00	$0.94 + 2 \times 0.51 = 1.96$	$1.07 + 2 \times 0.33 = 1.73$
Phosphorus = 6.00	$0.96 + 2 \times 0.67 = 2.30$	$1.07 + 2 \times 0.59 = 2.25$

“These calculations are for obvious reasons somewhat uncertain, especially because it is difficult to know if a constituent is an impurity or if it belongs to the structure. However, it is evident that in these cases the oxygen atoms have the necessary charges to compensate the missing univalent anions.

“If the oxygen atoms occupy the fluorine positions some of them will be unoccupied and the surrounding CaII atoms consequently uncompensated. If, on the other hand, the oxygen atoms occupy one half of the chlorine positions of the channels, all CaII atoms will be compensated, but the distribution around them will be rather unsymmetrical because the oxygen atoms will only occupy one position in each unit cell with the other one empty. It might be remarked that if the inter-atomic distances within the apatite structure are unchanged the distance channel-oxygen-calcium II will be rather large in this last case.

Table 21.

1	2	3	4	5	6
K ₂ O	0.07	K 0.02	9.90	10.00	10.07
Na ₂ O	0.03	Na 0.00			
CaO	52.00	Ca 9.27			
MnO	4.31	Mn 0.61			
FeO	0.00	Fe ²⁺ 0.00	5.90	5.96	6.00
MgO	—				
Li ₂ O	0.01				
Al ₂ O ₃	0.15				
Fe ₂ O ₃	0.35		0.93	0.94	0.96
P ₂ O ₅	41.84	P 5.90			
SiO ₂	—	F 0.75			
F	1.43	Cl 0.00			
CO ₂	0.00	OH 0.18	24.26	24.51	24.67
H ₂ O ⁺	0.17	O 24.26			
H ₂ O ⁻	0.04				
Insol.	0.18				
	100.70				
less for F, Cl	0.60				
	100.10				

- 1: Manganvoelckerite, Varuträsk (analyst Th. Berggren) after P. Quensel (1937).
- 2: Number of molecules in unit cell, density 3.21. Unit cell volume 516 Å³.
- 3: Number of atoms in unit cell.
- 4: Number of atoms if large cations = 10.00.
- 5: Number of atoms if phosphorus = 6.00.
- 6: Ideal number of atoms in the apatite structure.

Table 22.

	1	2	3	4	5	6
K ₂ O	—	—	K	—		
Na ₂ O	0.26	0.04	Na	0.08		
CaO	47.66	8.63	Ca	8.62		
MnO	6.65	0.95	Mn	0.95	9.97	10.00
FeO	2.32	0.32	Fe ²⁺	0.32		
MgO	0.04	—	Mg	—		
Li ₂ O	0.1	—	Li	—		
Al ₂ O ₃	—	—	Al	—		
Fe ₂ O ₃	—	—	Fe ³⁺	—		
P ₂ O ₅	41.46	2.96	P	5.92	5.92	6.00
SiO ₂	0.00	—	Si	—		
F	1.56	0.83	F	0.83		
Cl	0.50	0.14	Cl	0.14	1.07	1.08
CO ₂	0.00	—				2.00
H ₂ O ⁺	0.10	0.05	OH	0.10		
H ₂ O ⁻	0.00	—	O	24.26	24.26	24.59
Insol. . . .	0.04	—				24.00
	100.59					
less for F, Cl	0.66	-0.42				
	99.93					

1: "Mangualdite". Mangualde (analyst Th. Berggren) after B. Mason (1941).

2: Number of molecules in unit cell, density 3.28. Unit cell volume 313 Å³.

3: Number of atoms in unit cell.

4: Number of atoms if large cations = 10.00.

5: Number of atoms if phosphorus = 6.00.

6: Ideal number of atoms in the apatite structure.

"The first possibility seems to be quite improbable, but the second might explain why the pure compound has never been observed and why it seems to be difficult to synthesize the compound and why it is very sensitive to weathering.

"Can this hypothesis be tested? The only possibility seems to be a comparison of the X-ray intensities of critical reflections. Unfortunately it is not possible to study single crystals, and therefore the discussion must be based on the powder data. A calculation shows that it is not possible to distinguish with any degree of certainty on a powder diagram between the possible arrangements of the channel oxygen.

"One small point can be settled however. As mentioned the distance Ca II-oxygen will be rather large (2.9 Å) and it might be suspected that the calcium atoms around the channels will be closer together than in the normal apatite structure. An investigation shows that this is possible without any replacement of the phosphate group. The inter-atomic distances between the calcium atoms decrease very rapidly however and the expected displacement must therefore be rather small.

"If the parameter of the CaII atoms is varied a calculation shows that on a powder diagram the reflection 30 $\bar{3}$ 0 will be much weaker than the reflection 11 $\bar{2}$ 2 if the CaII-oxygen distance is decreased. Now it is found that on a powder diagram these two reflections have the same intensities as in an ordinary apatite mineral of the same manganese content. Thus it can be concluded that the parameters of the calcium atoms as well as the majority of the other atoms have the same positions as in an ordinary apatite.

"The voelckerite problem is not easy to solve because the effects on the diffracted X-rays are very small. On the other hand the evidence of the chemical analyses,

the weathering properties and the colour cannot be neglected and therefore I think we have reasonably good evidence for accepting the mineral manganvoelckerite."

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- MASON, B. (1941), Mangualdite is manganvoelckerite. *G.F.F.* 63, 383-386.
- MASON, B. (1941 a), Minerals of the Varuträsk pegmatite. XXVI. Mangan-hydrooxyapatite. *G.F.F.* 63, 279-284.
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- ROGERS, A. F. (1912), Dahilite (podolite) from Tonopah, Nevada; Voelckerite, a new basic calcium phosphate; remarks on the chemical composition of apatite and phosphate rock. *Amer. J. Sci.* 33, 475-482.
- WICKMAN, F. E. (1954), Manganvoelckerite. Minerals of the Varuträsk pegmatite XXXVIII. *G.F.F.* 76, p. 495.

20. Vivianite

Summarized by BRIAN MASON

"Vivianite has been observed as a thin coating on the iron-manganese phosphates triplite and triphylite. The amount of this mineral is minute, but the prominent blue colour makes it readily recognisable. The identification has been checked by optical tests. A small amount was scraped from the surface of triplite; the optical properties agreed with those of vivianite, the mineral being highly birefringent with $\alpha = 1.58$, $\gamma = 1.63$."

REFERENCE

- MASON, B. (1941), Minerals of the Varuträsk Pegmatite XXVII. *G.F.F.* 63, p. 285.

21. Beryl

Like several other minerals of the Varuträsk pegmatite, beryl occurs in differing aspects, representing different phases of mineralisation.

Around the prospecting pits F and G in the western wing the mineral is most abundant, often occurring in large coarse crystals, up to 30 cm in length. The beryls of this type are milky white on fresh fractures. The refractive indices are $\omega = 1.583$, $\varepsilon = 1.577$.

Another type shows a different aspect. In contrast to the well-defined crystals of the former type, the mineral now occurs in the form of rounded fragments of 2-3 cm in diameter, enclosed in a massive lepidolitic rock described below (p. 70). This beryl is pure white with a vitreous lustre. A third also vitreous type is found enclosed in spodumene, pictured in Fig. 55 (p. 102).

Analyses by Miss Berggren of these two types are given in Table 23. Chemically they are much alike. The somewhat divergent content of lithium and caesium may be accounted for by the mineral occurring in different assemblages.

A single crystal of beryl, dark brown in colour and in habit wholly diverging from the types referred to above, has been found. Unfortunately the locality within the pegmatite from which this crystal originated is not recorded, as it was collected amongst dumped matter. A spectrographic analysis, executed by Mr. Danielsson of the Boliden Company, showed a content of 2.8 % Cs_2O . This represents a caesium beryl with near the same content of Cs_2O as the species named Worobieffite by Vernadsky (with 3.10 % Cs_2O).

Table 23.

	1	2		1	2
SiO ₂	63.98	64.16	Li ₂ O	0.36	0.39
TiO ₂	0.00	0.00	Na ₂ O	1.09	1.27
Al ₂ O ₃	18.83	18.73	K ₂ O	0.16	0.08
Fe ₂ O ₃	0.16	0.28	Rb ₂ O	0.00	0.00
BeO	12.87	12.98	Cs ₂ O	0.26	0.42
MnO	0.01	0.00	H ₂ O ⁺	2.00	1.44
CaO	0.00	0.00	H ₂ O ⁻	0.05	0.02
MgO	0.00	0.00			
Sp. gr.				99.77	99.77
				2.712	2.725

1: Beryl, Varuträsk. Large massive crystal. Excavation F.

2: Beryl, Varuträsk. Inclusion in lepidolitic rock, quarry H.

REFERENCES

VERNADSKY, W. J. (1908), *Zeitschr. f. Kryst.* 50, p. 73.QUENSEL, P. (1937), *Minerals of the Varuträsk Pegmatite VII. G.F.F.* 59, p. 269.

22. The tourmaline group

Both black and coloured tourmalines are of frequent occurrence in the Varuträsk pegmatite. The former are principally restricted to the wall zone. A sketch map by

Table 24.

	1	2	3	4
SiO ₂	35.76	37.30	38.10	38.06
TiO ₂	0.25	0.06	0.09	0.02
Al ₂ O ₃	34.76	37.63	38.50	41.78
Fe ₂ O ₃	—	—	—	0.035
FeO	12.99	4.53	3.87	—
MnO	0.33	1.04	0.46	0.45
CaO	0.00	0.18	0.10	0.72
MgO	0.58	0.00	0.10	0.02
Li ₂ O	0.11	1.05	1.52	1.55
Na ₂ O	3.06	4.24	2.93	2.48
K ₂ O	0.06	0.10	0.21	0.25
Rb ₂ O	0.00	0.00	0.00	0.01
Cs ₂ O	0.00	0.00	0.00	0.00
B ₂ O ₃	10.00	10.18	10.70	10.88
F	0.37	0.77	0.77	0.92
P ₂ O ₅	—	—	—	—
Cr ₂ O ₃	—	0.00	0.00	0.00
Cu.Ni.Co	—	0.00	—	—
H ₂ O ⁺	1.93	2.80	2.69	3.02
H ₂ O ⁻	0.01	0.00	0.00	0.00
	100.21	99.88	100.04	100.195
O for F	0.16	0.32	0.32	0.39
	100.05	99.56	99.72	99.805
Sp. gr.	3.178	3.110	3.065	3.025

1: Black tourmaline (schorlite) Varuträsk (all Fe calculated as FeO).

2: Blue tourmaline (indicolite) Varuträsk (all Fe calculated as FeO).

3: Green tourmaline (verdelite) Varuträsk (all Fe calculated as FeO).

4: Red tourmaline (rubellite) Varuträsk.

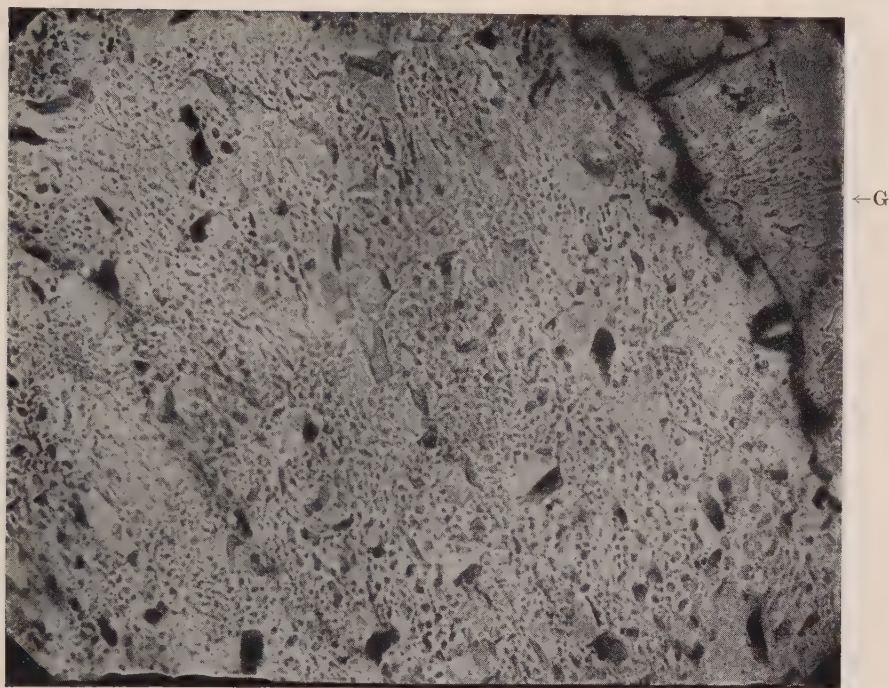


Fig. 27. Core of rubellite in zonal tourmaline, completely replaced by a quartz-albite intergrowth. G indicates the outward zone of green tourmaline. $\times 180$. Photo S. Hedlund.

O. Gabrielsson, produced in the original description (Quensel, P. and Gabrielsson, O. 1939, p. 64), gives a good general view of the field distribution of both the black and the coloured tourmalines.

Of the coloured species, the green verdelite is the most common and has the widest distribution in different phases of mineralisation in the pegmatite.

Rubellite is principally restricted to the sodium replacement unit being found there occasionally in large radiating groups. In one instance only it has been found participating in the mineral assemblage of the preceding caesium replacement unit, where it occurs in large crystals encompassed in a vitreous pollucite (Pl. I). The blue indicolite is relatively scarce at Varuträsk.

Zonal tourmalines are common in the sodium replacement unit. Without exception the outward zone consists of a thin shell of vivid light-green verdelite, encompassing a core of rose-coloured rubellite. The contact between the shell and the core is sharply defined, the outer shell readily peeling off, leaving the core intact.

The refractive indices are:

Black tourmaline (schorlite)	$\epsilon = 1.6324$,	$\omega = 1.6607$
Green tourmaline (verdelite)	$\epsilon = 1.6224$,	$\omega = 1.6423$
Red tourmaline (rubellite)	$\epsilon = 1.6205$,	$\omega = 1.6391$
Blue tourmaline (indicolite)	$\epsilon = 1.6230$,	$\omega = 1.6427$

Four analyses by Miss Berggren including the four different modifications are given in Table 24.

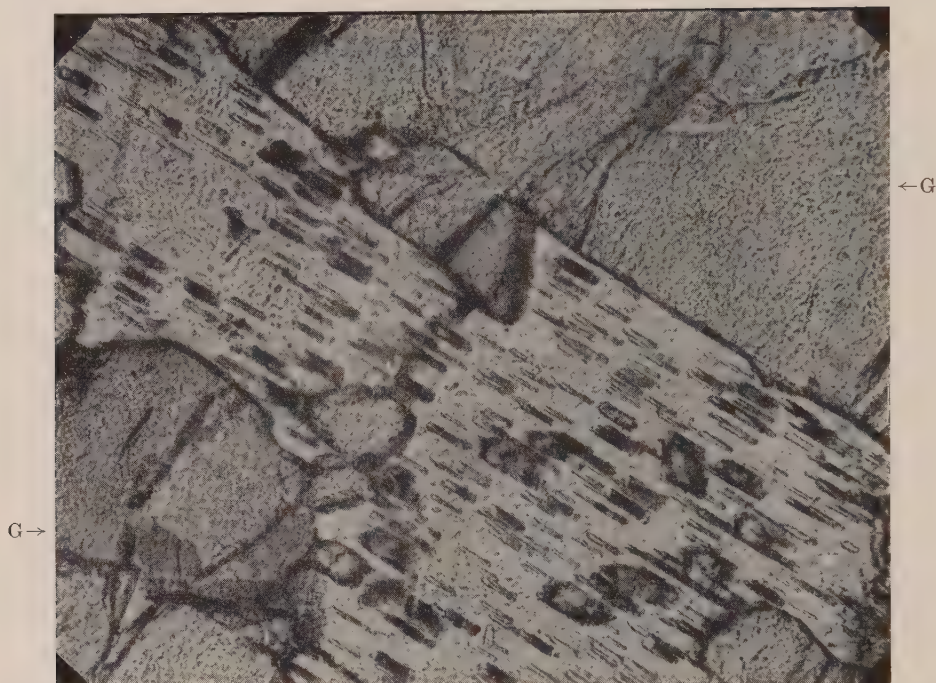


Fig. 28. Zonal tourmaline. Relics of core rubellite retaining a uniform orientation within the replacing quartz-albite intergrowth. An outer zone of green tourmaline (G) has remained unaffected. $\times 180$. Photo S. Hedlund.

Gabrielsson has compared the chemical composition of these tourmalines with those from a number of other localities. Correspondence between chemical composition, optical determinations and density are elucidated in several diagrams (cf. *l.c.*, pp. 75–84).

Of all the different tourmalines at Varuträsk only the red modification has been found to succumb to alterations. These occur in several different forms. In the first place may be mentioned complete pseudomorphs of cookeite. A fine-grained lepidolitic mica may also replace rubellite.

It is, however, the red cores of the zonal tourmalines which offer the most interesting forms of alteration. In many such cases the red core is more or less completely replaced by a symplectitic intergrowth of albite and quartz (Fig. 27). The green shell then remains intact. Under the microscope one can often observe innumerable small relicts of the rubellite, retaining a uniform optical orientation (Fig. 28). Occasionally the remaining portions of the rubellite may be preserved as a complete framework of orientated microlites, embedded in the delicately textured albite-quartz-symplektite. The import of the alterations will be considered in the paragenetic part of this paper (p. 113). The core may also be completely replaced by cookeite and spodumen (cf. p. 72).

REFERENCE

QUENSEL, P. and GABRIELSSON, O. (1939), Minerals of the Varuträsk Pegmatite XIV. G.F.F. 61, p. 63.



Fig. 29. Vein of second generation of spodumene in microcline perthite. Upper part unaltered, lower part, below dotted line, completely decomposed to clay minerals.

23. Spodumene

During the first years of mining at Varuträsk spodumene was only found in small quantities. During more extended quarrying later, however, large quantities of the mineral were excavated.

Much of the spodumene is developed in the form of thick tabular masses, highly intercalated with quartz. When recurring in a younger epoch of mineral deposition the habit has changed to accumulated masses of elongated slender laths. (Cf. Figs. 44 and 53, pp. 90, 100.)

The refractive indices of the first type are $\alpha = 1.661$, $\beta = 1.666$, $\gamma = 1.676$.

Two analyses were made by Miss Berggren. No. 1 in Table 25 is of the younger type. No. 2 is of a semi-transparent wine-yellow type, only found in a few specimens.

The spodumene at Varuträsk frequently succumbs to alterations. Locally it is found to be replaced by cookeite. More frequent is an alteration to kaolinite. The identity of kaolinite (*sensu stricto*) has been verified by X-ray analysis. No. 3 in the Table 25 gives an analysis by Miss Berggren thereof. The refractive indices are $\alpha = 1.543$, $\gamma = 1.556$.

In other cases the alteration products of spodumene have a more complicated composition, consisting of a mixture of different clay minerals. In Fig. 29 a large vein of spodumene, belonging to the younger generation, is in its lower part, from the dotted line downwards, completely altered to a softclay-like substance, readily

Table 25.

	1	2	3
SiO ₂	64.34	64.89	44.08
TiO ₂	0.00	0.00	0.00
Al ₂ O ₃	27.01	26.74	33.77
Fe ₂ O ₃	0.24	0.57	1.27
FeO	0.00	0.04	1.11
MnO	0.01	0.01	0.02
CaO	0.00	0.00	0.50
MgO	0.00	0.00	1.79
Li ₂ O	7.00	7.12	0.53
Na ₂ O	1.04	0.05	0.27
K ₂ O	0.08	0.16	0.88
P ₂ O ₅			0.36
H ₂ O ⁺	0.24	0.48	11.94
H ₂ O ⁻	0.07	0.06	3.32
	100.03	100.12	99.84
Sp. gr.	3.130	3.163	2.533

1: Spodumene.

2: Spodumene, semi-translucent wine-yellow type.

3: Kaolinite as alteration of spodumene (rotten spodumene).

scooped out by hand. In this case minerals of the montmorillonite group also participate in the alteration products (cf. p. 114).

REFERENCES

QUENSEL, P. (1938), Minerals of the Varuträsk Pegmatite X. G.F.F. 60, p. 201.

ALVFELDT, O. (1941), Minerals of the Varuträsk Pegmatite XXII, G.F.F. 63, p. 49.

24. The mica group

Miss Berggren undertook a chemical investigation of all the micas of the Varuträsk pegmatite (Th. Berggren 1940, 1941). Miss Lundblad has determined the optical properties of most of the analysed samples (B. Lundblad 1942).

In recalculating the analyses of the micas, Miss Berggren has followed Stevens in assuming that lepidolite as a mineral species is an isomorphous mixture of polyolithionite, lithiummuscovite, muscovite and biotite radicals (R. E. Stevens 1938). The unit cell content for the four radicals is taken to be:

Polyolithionite	K ₄ Li ₈ Al ₄ Si ₁₆ O ₄₀ F ₈
Lithiummuscovite	K ₄ Li ₆ Al ₆ Al ₄ Si ₁₂ O ₄₀ (OH,F) ₈
Muscovite	K ₄ Al ₈ Al ₄ Si ₁₂ O ₄₀ (OH,F) ₈
Biotite	K ₄ Mg ₁₂ Al ₄ Si ₁₂ O ₄₀ (OH,F) ₈

Fig. 30. Composition of the mica minerals of the Varuträsk pegmatite according to Thelma Berggren (Berggren 1941).

The numbers refer to those in the table of analyses, No. 26.

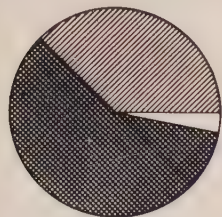
For a comparison with the original numbers the connections are given below:

Berggren's designations: A, B, C, D, E, F, G, H, J, K, L

Numbers in Table 26: 1, 4, 5, 6, 7, 10, 13, 11, 12, 9, 8

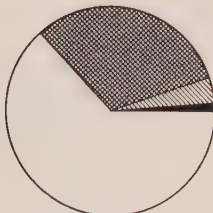
Numbers 2 and 3 are later analyses, not included in Berggren's original compilation. They represent Nos. 10 and 11 published in B. Lundblad's paper (l.c.). No. 3 is recalculated after deduction of free quartz (cf. p. 71).

Polyolithionite Series

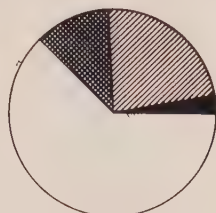


1

Lithiummuscovite Series

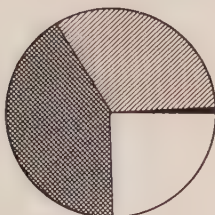


2 Large crystals of lepidolite

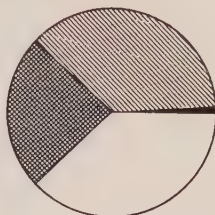


3 Gray lepidolite

Lepidolite Series

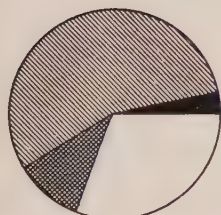


4 Colourless lep. Concentric

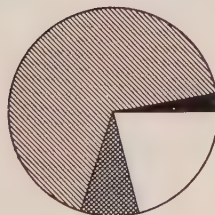


5 Fine-grained lepidolitic rock

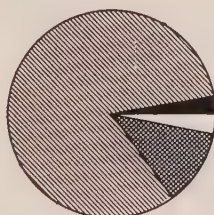
Muscovite Series



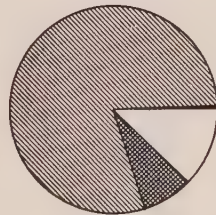
6 Gray lepidolite



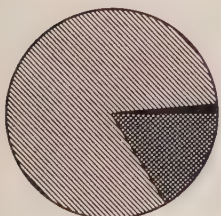
7 Gray lepidolite Concentric



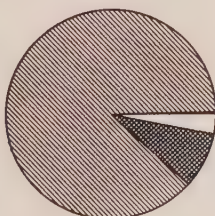
8 Muscovite Concentric



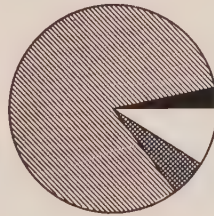
9 Sulphur-yellow muscovite



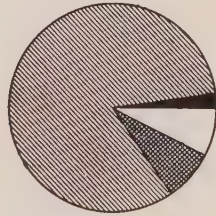
10 Purple oncosine



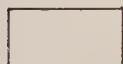
11 Gray oncosine



12 Muscovite



13 Rose-coloured muscovite



Lithiummuscovite



Polyolithionite



Muscovite



Biotite

Fig. 30.

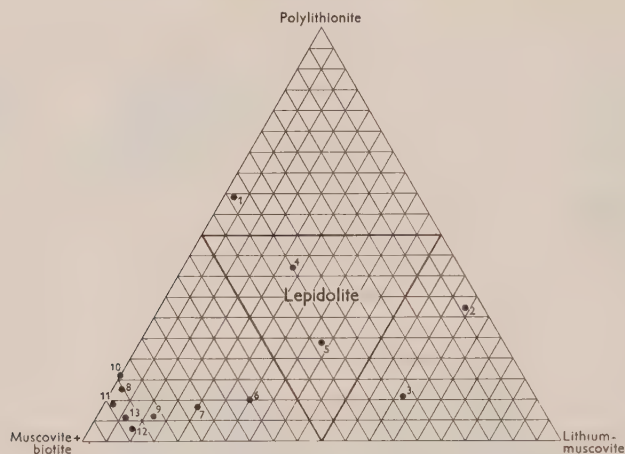


Fig. 31. Projected composition of the analysed mica minerals (Table 26) and represented in Fig. 30. (Numbers correspond to those of Table 26 and Fig. 30.)

Berggren proposed that the lepidolitic micas with more than 50 % of the polyolithionite radical be referred to a *polyolithionite series*, those with over 50 % lithium-muscovite radical to a *lithiummuscovite series*, and those with over 50 % muscovite radical to a *muscovite series*. Lepidolitic micas with contents below 50 % of any of the foregoing radicals, and consequently composing an isomorphous mixture of all three are referred to a *lepidolite series*. A graphic tabulation of this classification is given in Fig. 30. In a diagram (Fig. 31) the position of the different species is illustrated.

With the exception of the light gray lepidolite, pertaining to the polyolithionite series (Analysis 1) the analyses in Table 26 are grouped according to decreasing amount of Li_2O .

Levinson has recently studied the relations between polymorphism and chemical composition in micas of the muscovite-lepidolite series (A. A. Levinson 1953). Increasing amounts of lithium are found to conform with different layer structures. In Table 4 in his paper Levinson has calculated the layer structures for twelve of the analysed micas from Varuträsk. Of these 9 micas show normal muscovite structure (Analyses 5, 6, 7, 8, 9, 11, 12 and 13 in Table 26 in this paper). Two micas show one-layer lepidolitic structure (Analyses 2 and 4) and one six-layer lepidolitic structure (Analysis 1).

Levinson says that the analysed micas from Varuträsk do not fit well with his "idealised representation of relationship between Li_2O content and polymorphism in the muscovite-lepidolite series" (Levinson, l.c., Fig. 5). He assumes that this may be due to the circumstance that several of the Varuträsk micas are optically non-homogeneous. Lundblad has shown this to be the case with the micas of analyses 2, 3 and 7 (see data given below). A lepidolitic mica from Varuträsk, which Levinson found most inconsistent with the layer structure, contained 3.9 % Li_2O according to Miss Berggren's analysis (No. 5 in Table 26). As a check this mica was re-analysed by Heinrich who found 3.8 % Li_2O , in good agreement with Berggren's determination (Heinrich and other 1953, Table XII, p. 167). Levinson's supposition that the lithium content of the mica samples may vary so much, "that the portions supplied represent

Table 26. Analyses of the mica minerals of the Varuträsk Pegmatite. (Including optical determinations.)

	1	2	3	4	5	6	7	8	9	10	11	12	13
SiO ₂	54.05	49.80	49.09	50.00	48.68	46.34	46.30	46.24	46.96	47.64	46.17	45.18	46.01
TiO ₂	0.00	0.00	0.00	0.00	0.00	0.06	0.00	0.10	0.06	0.00	0.00	0.15	0.00
Al ₂ O ₃	22.88	25.56	25.50	24.96	29.62	32.47	33.08	32.37	36.48	34.22	35.57	35.76	35.64
Fe ₂ O ₃	0.17	0.08	1.04	0.24	0.10	0.00	0.00	0.00	0.43	0.10	0.15	0.00	0.13
FeO	0.00	0.00	0.57	0.00	0.00	1.06	1.20	1.14	0.15	—	0.08	1.52	0.00
MnO	0.00	0.38	0.51	0.16	0.28	0.35	0.28	0.09	0.01	0.05	0.04	0.11	0.09
CaO	0.00	0.00	0.00	0.20	0.00	0.36	0.00	0.10	0.00	0.00	0.00	0.00	1.12
MgO	0.00	0.22	trace	0.01	0.00	0.00	0.14	0.19	trace	0.28	0.00	0.07	0.04
Li ₂ O	4.55	5.95	5.08	4.35	3.9	2.45	1.80	1.1	1.10	1.10	0.76	0.73	0.69
Na ₂ O	0.59	0.4	0.57	0.67	1.3	0.5	0.63	0.79	0.65	0.47	0.56	0.88	1.88
K ₂ O	10.17	9.67	11.77	9.95	10.06	9.46	10.09	10.16	8.69	10.40	10.37	9.95	8.19
Rb ₂ O	2.11	1.97	1.85	2.70	1.5	1.5	1.37	1.3	1.0	0.35	1.1	0.57	1.20
Cs ₂ O	1.25	1.2	0.33	1.90	0.2	0.2	0.41	0.2	0.1	0.75	0.3	trace	0.20
F	5.67	6.85	3.70	4.95	4.60	2.82	2.06	1.41	0.51	1.21	0.76	0.88	0.54
Cl	—	—	—	—	—	—	—	—	—	—	—	—	—
H ₂ O ⁺	0.82	0.50	1.13	1.30	1.64	3.32	3.06	3.41	4.08	3.62	4.06	4.48	4.65
H ₂ O ⁻	0.09	0.38	0.06	0.60	0.14	0.32	0.34	0.69	0.09	0.10	0.12	0.38	0.08
less O for F	102.35	102.96	101.20	101.99	102.02	101.21	100.76	100.63	100.31	100.29	100.04	100.66	100.46
	2.39	2.89	1.45	2.08	1.94	1.19	0.87	0.59	21	0.51	0.32	0.37	0.23
	99.96	100.07	99.75	99.91	100.08	100.02	99.89	100.06	100.10	99.78	99.72	100.29	100.23
Sp. gr.	—	2.898	—	—	2.831	2.865	2.803	—	—	—	—	2.768	2.865
2V _{Na}	—	36°	—	50°	36°	43°	43°	43°	44°	—	—	42°	—
2E	—	58°25'	—	—	—	—	69°7'	68°17'	—	—	—	66°52'	—
n	—	1.57	—	—	—	—	1.56	1.54	—	—	—	1.54	—

1. Light gray lepidolite (polyolithionite series)
2. Large crystals of lepidolite (lithium-muscovite series)
3. Gray lepidolite in veins (lithium-muscovite series)
4. Colourless to light rose lepidolite (lepidolite series)
5. Lepidolite in fine-grained lepidolitic rock (lepidolite series)
6. Gray lepidolite (muscovite series)
7. Gray lepidolite, concentric (muscovite series)
8. Muscovite, concentric (muscovite series)
9. Sulphur-yellow muscovite, concentric (muscovite series)
10. Cryptocrystalline muscovite in veins (muscovite series)
11. Cryptocrystalline muscovite (muscovite series)
12. Muscovite in normal development (muscovite series)
13. Rose-coloured muscovite (muscovite series)

extreme structural variations of the analysed material" does not seem to be consistent with the practically constant amount of lithium, found by Berggren and in the re-analysed sample by Heinrich. If, as Levinson anticipates, an inconsistency exists in combining micas with 3.8–3.9 % Li_2O with normal muscovite layer structure, influences of another nature than only the lithium ratio must probably be taken into consideration. The not insignificant amount of rubidium and caesium in most of the lepidolitic micas in the Varuträsk pegmatite may on account of their larger atomic dimensions influence their applicability to different layer structures.

The different mineral species of the mica group in the Varuträsk pegmatite offer many dissimilarities in general aspect. As these will be commented on in the paragenetical section of this paper on account of their characteristic affinity to different phases of mineralisation, a short review of the most conspicuous modifications may suffice in this connection.

Only one species can be classified as pertaining to the polyolithionite series (Table 26, Analysis No. 1). It is only found within a restricted area, associated with the small mass of pollucite around quarry F. This mica is light gray in colour with a resplendent lustre. Determinations with the universal stage showed that it is optically heterogeneous, consisting of one component with $2V_{\text{Na}} 34^\circ$ and another with $2V_{\text{Na}} 48^\circ$. In this sections one can easily distinguish the two component from one another (cf. Lundblad 1942, p. 56).

Another species worth attention is of a very different aspect. It forms large tooth-shaped crystals up to 7 cm in length and 2 cm in diameter often with a well-defined hexagonal boundary (Fig. 32). This type is principally found around a small prospecting pit T. The content of 5.95 % Li_2O is the highest in any of the analysed micas from Varuträsk (Analysis 2) and also the highest recorded in the 29 analyses of lepidolite in Doelter's *Handbuch der Mineralchemie*.

The greatest accumulation of lepidolite is concentrated in a fine-grained lepidolitic rock mass in the southern part of the western wing. Many hundred tons of it have been excavated. (Analysis 5.)

Two of the analysed mica minerals form a distinct group, both in general aspect and in mode of occurrence. Both are developed as dense cryptocrystalline aggregates, indicated by applying the name oncosine to them as ascribed by Tschermak for a "dichter Muscovite" (Tschermak, *Lehrb. der Min.* 1915, p. 633).

In one case this muscovite occurs as veins in pollucite (cf. p. 75 and Fig. 35). In other cases it is found in more independent masses. In chemical composition the two occurrences are much alike (Analyses 10 and 11). The vein material is purple, the more massive type grayish in colouring.

The gray micas (Analyses 3, 6 and 7) form a characteristic group, representing a late phase of mineralisation, commented on the paragenetical description (p. 107). They occur both as vein material as well as in accumulated masses.

The vein type has a widespread distribution, traversing practically all other mineral assemblages of the pneumatogenic stage. It was found to be optically heterogeneous. The greater part is uniaxial, other parts are distinctly biaxial with a large optical angle ($2V_{\text{Na}}$ about 42°). In boundary zones between the two modifications one can follow how the optical angle successively opens. Hendricks and Jefferson describe a similar condition in lepidolite from Londonderry (Western Australia) as follows: "The sheet is predominantly uniaxial but in parts the optic axis opens up to 20° – 40° $2V$ " (S. B. Hendricks and M. E. Jefferson 1939). The analysis of this type is No. 3 in Table 26. The material could not be freed from finely disseminated

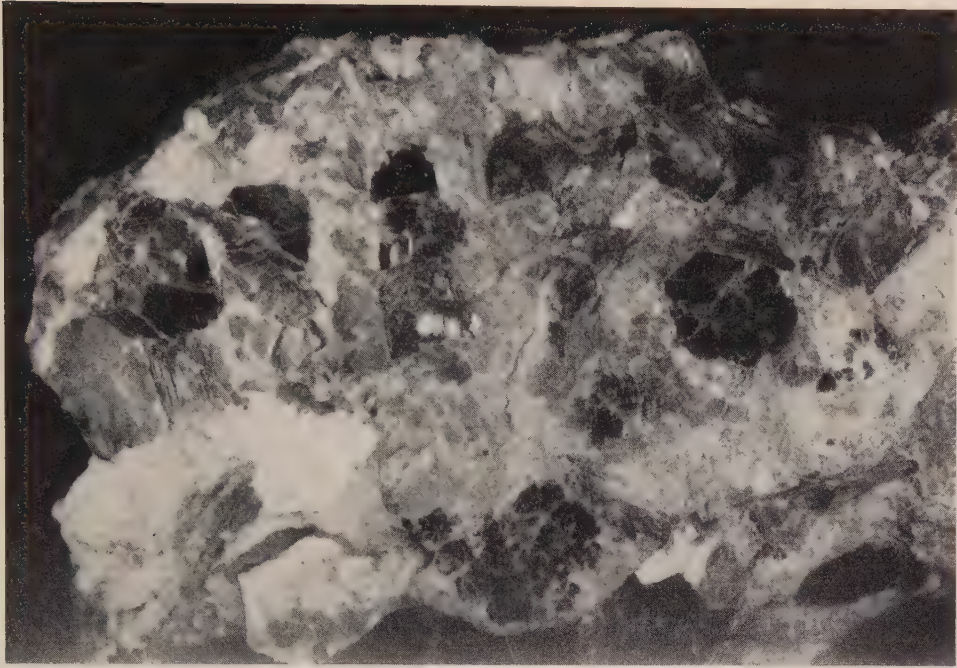


Fig. 32. Large toothshaped crystals of lepidolite in cleavelandite. Excavation T. Nat. size.
Photo O. Mellis.

quartz. The analysis has therefore been recalculated on an estimated percentage of 25.50 % Al_2O_3 . The result is so well in accordance with the general composition of the lepidolitic micas in Varuträsk that the recalculation can be considered approximately satisfactory.

A gray mica occurring in concentric bundles is also optically non-homogeneous. Different parts of even the same cleavage plate gives values for $2V_{\text{Na}}$ varying between 32° and 43° (Analysis No. 7 in Table 26).

The frequent occurrence of rose-coloured micas at Varuträsk may be noted as often recorded in other lithium pegmatites (cf. E. Wm. Heinrich and A. A. Levinson 1943). One type, forming concentric bundles, is a true lepidolite with 4.35 % Li_2O (Analysis No. 4). The other analysed type (Analysis 11) is a very fine-grained muscovite with only 0.69 % Li_2O .

Finally the abundant occurrence of muscovite in normal platy development (Analysis 13) or with a tendency to develop a concentric structure (Analysis 8) may be mentioned. The former is the dominating mica of the wall- and intermediate zones of the pegmatite stage. Both the analysed samples contain a certain amount of lithium (0.73 and 1.1 % Li_2O resp.).

Many instances pertaining to the general aspects of the different mica minerals will, as already noted, be considered in connection with their paragenetical participation under a later heading. Further details in this connection may therefore be superfluous.

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25. Cookeite

Under a previous heading cookeite and spodumene have been noted as replacing rubellite in the zonal tourmalines. Cookeite is, however, also found to occur as a self-existent phase of mineralisation, filling out cracks and cleavages irrespective of any special mineral association.

The cookeite is in all occurrences developed in very fine-grained and compact masses. With the naked eye no features are observable. Its colour may vary from light yellowish-green to light pink.

Under the microscope the mineral is seen to form a dense, cryptocrystalline network of scaly individuals. The refractive indices for the analysed sample are $\alpha = 1.565$, $\gamma = 1.595$.

An analysis of cookeite, pseudomorph after rubellite, is given in Table 28. The somewhat high content of SiO_2 is due to a small content of quartz in the analysed specimen.

The principal change in chemical composition between the red tourmaline and the cookeite replacing the same is the complete removal of boron and an addition of lithium and water. The content of Li_2O has increased from about 1.45 % in the rubellite (Analysis No. 4 in Table 24) to 4.33 % in the replacing cookeite.

The formation of cookeite during the subsequent mineralisation of the Varuträsk pegmatite will in the paragenetic division of this paper be referred to the hydrotogene stage. The lower temperature then prevailing would favour the formation of cookeite rather than the less hydrous lepidolitic micas (cf. p. 113).

Table 28.

	1	2		1	2
SiO_2	38.22	35.09	K_2O	0.42	
TiO_2	0.00		P_2O_5	0.11	
Al_2O_3	43.20	44.74	F	0.33	
Fe_2O_3	0.08		Cl	0.03	
FeO	0.07		CO_2	0.00	
MnO	0.03		SO_3	0.00	
CaO	0.36		H_2O^+	12.46	15.79
MgO	0.04		H_2O^-	0.16	
Li_2O	4.33	4.38			
Na_2O	0.00			99.84	
Sp. gr.					2.68

1: Cookeite, replacing rubellite as complete pseudomorph.

2: Theoretical composition according to the formula $\text{Li Al (OH)}_{23} \cdot \text{Si}_2\text{O}_6$.

REFERENCE

- QUENSEL, P. (1937), Minerrals of the Varuträsk Pegmatite VI. G.F.F. 59, p. 262.

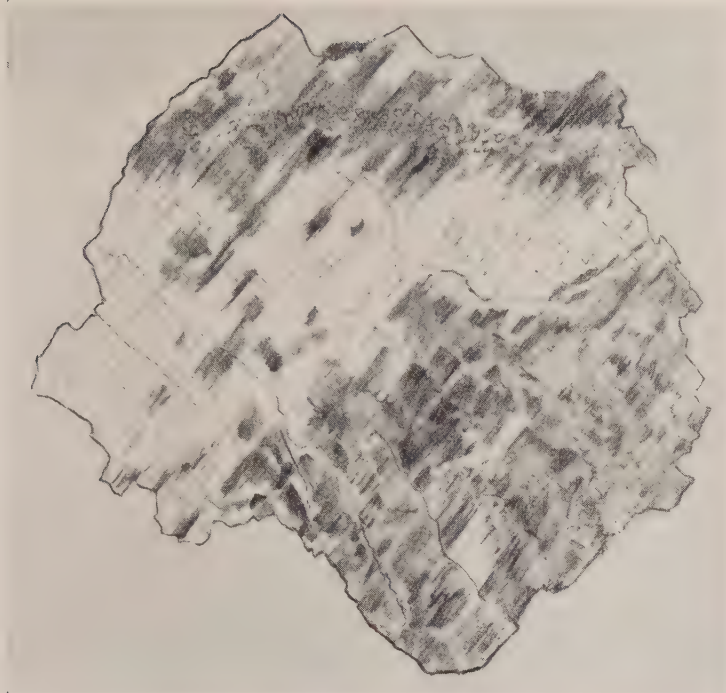


Fig. 33. Incipient alteration of petalite (light) to montmorillonite (dark) along cleavage planes.
 × 15. M. Rosenberg del.

26. Petalite

Petalite occurs in large masses in the western wing of the pegmatite. Around quarry D some thousand tons of the mineral have been excavated.

The petalite is developed in columnar masses or in large well cleavable platy bodies. Crystal faces are not developed. When fresh the mineral is grayish in colour with a vitreous, on cleavage planes pearly, lustre. It is in general sub-translucent, on cleavage planes sub-transparent to transparent, comparable with the sub-transparent petalite from Londonderry (West Australia) though not attaining the gem variety from that locality.

The cleavage planes on (001) are perfect, on (201) imperfect. An analysis by Miss Berggren is given in Table 29.

In Europe petalite has, as far as known, only been recorded from seven other localities. Besides the original occurrence in Elba, it is found in the Utö pegmatite (Sweden) at Kuivula in Finland, at Oakhampton and in a pegmatite related to the Dartmoor granite (personal communication) in England as well as at Jeclov near Jihlava (Moravia) and at a locality near the source of the Amannus river, Batalpaschinsk (Northern Caucasus).

The petalite at Varuträsk readily succumbs to an alteration into montmorillonite. In an initial stage it is confined to the cleavage planes or to the terminal ends of crystalline groups (Fig. 33). The montmorillonite is then fibrous. On further decompo-

sition it becomes massive and earthy. The colour is pale pink, in larger masses deeper pink. Under the microscope a weak birefringence is observable. Mean index of refraction is 1.495. An analysis by Miss Berggren is given as No. 2 in Table 29.

REFERENCE

QUENSEL, P. (1937), Minerals of the Varuträsk Pegmatite IV, G.F.F. 59, p. 150.

Table 29.

	1	2		1	2
SiO ₂	77.18	44.72	Li ₂ O	4.11	0.00
TiO ₂	0.00	0.00	Na ₂ O	0.61	0.10
Al ₂ O ₃	17.05	21.69	K ₂ O	0.17	0.00
Fe ₂ O ₃	0.20	1.54	P ₂ O ₅	0.00	0.02
FeO			F	0.03	0.05
MnO	0.00	0.19	H ₂ O ⁺	0.44	24.32
CaO	0.14	2.83	H ₂ O ⁻	0.10	
MgO	0.00	4.54			
				100.03	100.00
Sp. gr.				2.418	

- 1: Petalite, Varuträsk. Th. Berggren anal.
2: Montmorillonite, alteration of petalite. Th. Berggren anal. Analysis recalculated to 100 after deduction of 1.69 % Li₂O as impurity of petalite.

27. Pollucite

The first specimens of pollucite were found by O. Ödman in 1938, five years after mining operations had begun in the pegmatite. Shortly afterwards considerable amounts of the mineral were located and during extended operations in the following years ever increasing quantities were exposed.

The largest masses are found around quarry P in the eastern wing of the pegmatite and in the underground workings from the shaft. In the western wing smaller quantities of the mineral have been found around location F.

The aspect of the pollucite from the two centres is somewhat different. In the eastern quarries the colour is grayish-white with a dull lustre (Fig. 34), whereas pollucite from the western wing is pure white and vitreous. Another difference is that all pollucite from the eastern wing is intersected by numerous veins of varying composition (cf. below) whereas nothing similar is found in the mineral from the western locality. Instead the dazzling white pollucite is there intergrown with large crystals of bright red tourmaline. The contrast in colour of the two minerals offers a striking aspect (see Pl. I).

Under the microscope the pollucite is fully isotropic. The indices of refraction, determined in monochromatic light, are $n_{Li} = 1.5190$, $n_{Na} = 1.5218$, $n_{Ta} = 1.5256$.

An analysis of pollucite from the eastern wing is given below (Table 30 No. 1). The ratios for SiO₂ and H₂O are somewhat high for the theoretical formula 2R₂O · Al₂O₃ · 9SiO₂ · H₂O, as is also the case in analyses of the mineral from other localities. This irregularity may according to Strunz be explained by (AlCs) in a limited degree being replaced by Si (H. Strunz, 1936). According to this supposition, Strunz computes the formula for pollucite to be (Si₃₂ Al₁₆ Cs₁₆ O₉₆ · 6–8 H₂O). The content of H₂O then needs to be fixed only approximately.

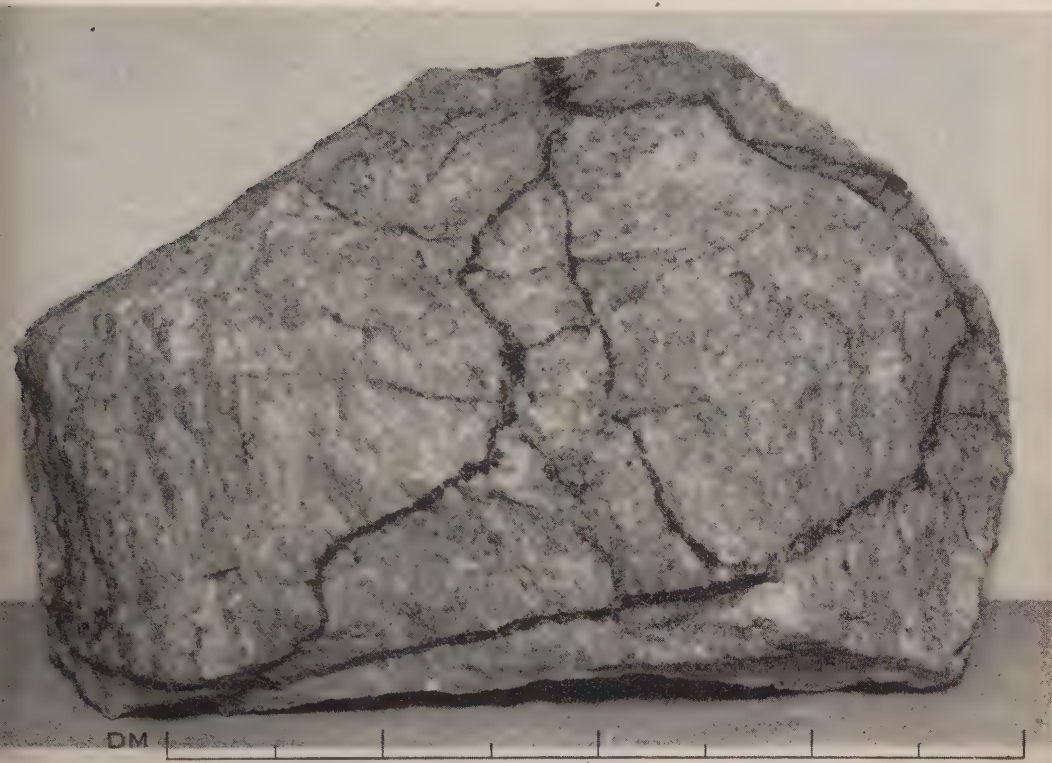


Fig. 34. Pollucite, intersected by veins of albite (dark). Quarry P. $\times \frac{1}{4}$. Weight of specimen 75 kg.
Photo S. Hedlund.

The most common of the numerous veins intersecting the pollucite in the eastern wing of the pegmatite are gray in colour. They vary in breadth from some mm up to 1.5 cm (see Fig. 34). These veins consist of almost pure albite.

A set of mauve-coloured veins comes next in importance. In width they have about the same variation as the albite veins. The material is dense, with the naked eye no details are perceptible (Fig. 35). Under the microscope, however, a distinct micaceous texture is observable. To verify the composition an analysis was made by Miss Berggren (Table 30, No. 10). The chemical composition is that of muscovite, though containing a relatively high content of both lithium and caesium. The cryptocrystalline texture of the mica was in the original description indicated by naming it oncosine.

A third group of veins, also abundantly intersecting the pollucite in the eastern wing, has a more complicated composition. These veins grade from microscopic dimensions up to some 4 mm in width. The colour is dull olive gray with a resinous lustre.

Under the microscope they are found to consist of spodumene in a fibrous development, irregularly orientated normal to the vein wall (Fig. 36). Higher magnification reveals that the spodumene is not uniform but composed of two phases in a finely dispersed intergrowth. (Fig. 37.)

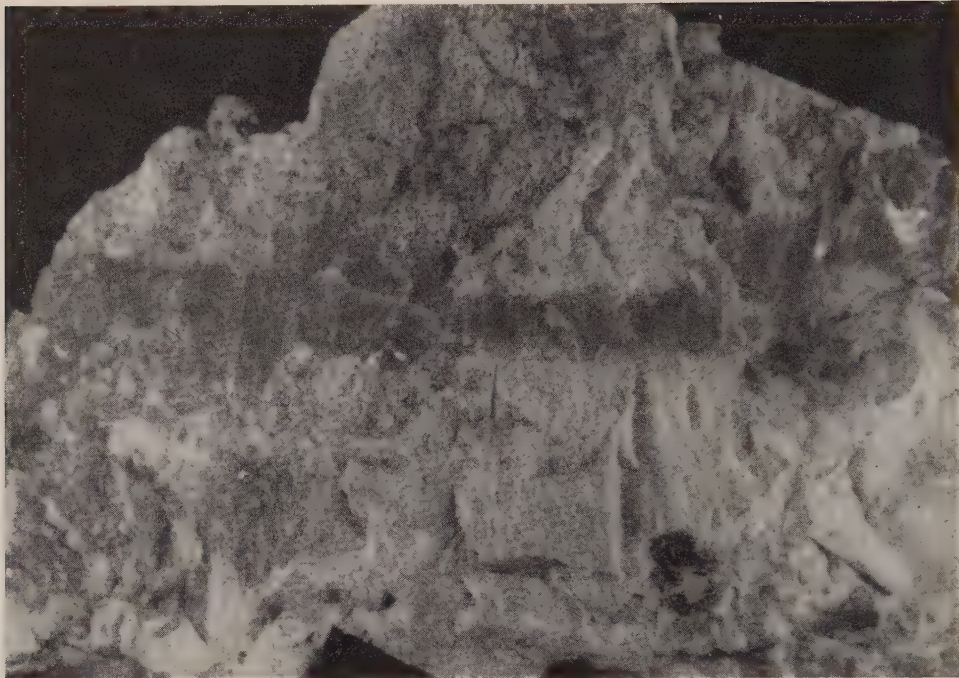


Fig. 35. Vein of cryptocrystalline muscovite (oncosine) in pollucite. Quarry P. Nat. size. Photo O. Mellis.

Table 30.

	1	2	3
SiO ₂	46.28	47.64	64.48
TiO ₂	0.00	—	—
Al ₂ O ₃	16.71	34.22	23.68
Fe ₂ O ₃	0.01	0.10	0.00
FeO	—	—	0.74
MnO	0.00	0.05	0.02
CaO	0.00	0.00	0.00
MgO	0.00	0.28	0.10
Li ₂ O	0.25	1.10	4.70
Na ₂ O	1.87	0.47	0.52
K ₂ O	0.51	10.40	0.84
Rb ₂ O	1.60	0.35	0.24
Cs ₂ O	30.77	0.75	3.73
Cl	0.00	—	—
F	0.03	1.21	—
P ₂ O ₅	0.25	—	—
H ₂ O ⁺	1.80	3.62	0.77
H ₂ O ⁻	0.00	0.10	0.05
	100.08	100.29	99.87
— O for F		0.51	
Sp. gr.	2.917	99.78	

- 1: Pollulite. Eastern quarry P.
2: Dense cryptocrystalline muscovite (oncosin). Vein in pollucite. (Cf. Table 26, analysis No. 10.)
3: Vein of spodumene in pollucite.



Fig. 36. Vein of spodumene in pollucite. $\times 12$. Photo O. Mellis.

Except for a somewhat high content of Al_2O_3 , the analysis of this vein material (No. 3, Table 30) corresponds approximately to the composition of spodumene. Optical determination on the universal stage gave $2V_{\text{Na}} = 58^\circ 30'$, $n = 1.67$, values well in accordance with those of spodumene. An X-ray examination by Professor Westgren gave similar results. The lattice dimensions were found to be the same as those given by Warren and Biscoe for spodumene (B. E. Warren, and J. Biscoe, 1931).

The nature of the other phase in the symplektite is however left unanswered. In the analysis nearly half of the content of Li_2O is replaced by Cs_2O . As the very fine-grained texture of the symplektite has evaded optical determinations of the second phase, it must for the present be left an open question in what way the content of caesium participates in the vein material. It cannot be in the form of inclosed grains of pollucite as the integrating component shows a definite though low birefringence. If it were not for the large difference in atomic radii of lithium and caesium one would be disposed to anticipate that a certain amount of caesium might have entered the spodumene lattice and later disintegrated to form the two components of the sym-



Fig. 37. Enlarged photograph of spodumene vein in Fig. 36, showing the symplektitic texture of the spodumene. $\times 120$. Photo O. Mellis.



Fig. 38. Second symplektitic intergrowth within the spodumene vein as seen in Figs. 36 and 37. $\times 160$, Nicols crossed. Photo S. Gavelin.

plektite. In any case the nature of a caesium component, not answering to pollucite, is unknown.

All that can be said at present is that the one phase of the symplektite, pictured in Fig. 37, is spodumene and the other phase may contain a content of caesium which in that case Westgren concludes "is probably too finely dispersed to give rise to observable X-ray interferences" (P. Quensel, l.c., p. 625).

In the same sections another symplektite intergrowth is found within the spodumene vein (Fig. 38). The minute dimensions of this intergrowth again impede reliable determinations. It has not been possible to select material for an analysis.

The low refractive indices and low birefringence of both components in this case exclude the participation of spodumene. The one component in all probability consists of feldspar, presumably albite. It has not been possible to attain any optical data indicative of the nature of the second component. Similarities within another occurrence of correlative nature may however offer some incidences worth consideration.

Though of a much finer texture this symplektite in the vein material at Varuträsk shows many aspects corresponding to what Brush and Dana have described as alterations of spodumene from Branchville (G. H. Brush and E. S. Dana 1880, p. 270). Under the microscope one can observe much the same textural configuration as pictured of correlative alterations of spodumene from the Branchville pegmatite where it is defined by the authors as an "albite-eucryptite mixture". This might lead to the supposition that the symplektite at Varuträsk also may consist of the same constituents. However, as exact determinations are lacking, a conclusive determination of the two phases, integrating in the symplektite, must await further investigation.

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- BRUSH, G. H. and DANA, E. S. (1880), *Am. J. of Sc.* 20, p. 270.
 QUENSEL, P. (1938), *Minerals of the Varuträsk Pegmatite XIII*. G.F.F. 60, p. 612.
 STRUNZ, H. (1936), *Zeitschr. für Krist.* 95, p. 1.
 WARREN, B. E. and BISCOE, J. (1931), *ibid.* 80, p. 391.

28. Albite

The only form of plagioclase in the pegmatite is all but pure albite, mostly developed as cleavelandite, often in the form of large spheroidal bursts (Fig. 39). When traversing other minerals in narrower veins it may be very fine-grained though under the microscope seen still to retain its cleavelandite habit.

Locally one may observe a transgression from such fine-grained cleavelandite to a sugar-grained albite. Such saccharoid albite has moreover a not insignificant distribution, though principally restricted to more peripheral parts of the pegmatite, presumably due to more rapid consolidation.

Three analyses have been made by Miss Berggren. Analysis No. 1 in Table 31, representing cleavelandite in normal development, contains no calcium (CaO 0.00 %). The saccharoid albite (No. 3) has only 0.03 % CaO . In both forms an exceptionally pure albite is therefore authenticated. This is further verified by optical determinations. Adamson found the extinction angle on (010) to be 20° , the axial angle $2V_\gamma = 77^\circ$.

Table 31.

	1	2	3
SiO ₂	67.61	69.03	68.30
TiO ₂	0.00	0.00	0.00
Al ₂ O ₃	20.17	18.97	19.64
Fe ₂ O ₃	0.03	0.06	0.08
FeO	0.00	0.02	0.00
MnO	0.001	0.01	0.00
CaO	0.00	0.26	0.03
MgO	0.00	0.00	0.00
Li ₂ O	0.036	0.04	0.004
Na ₂ O	11.65	10.78	11.65
K ₂ O	0.10	0.10	0.08
P ₂ O ₅	0.15	0.26	0.18
H ₂ O ⁺	0.14	0.25	0.08
H ₂ O ⁻	0.03	0.05	0.03
	99.92	99.83	100.07
Sp. gr.	2.620	2.599	2.612

1: Cleavelandite. Varuträsk (white).

2: Cleavelandite. Varuträsk (light rose-coloured).

3: Saccharoid-grained albite, Varuträsk.

REFERENCE

ADAMSON, O. J. (1942), Minerals of the Varuträsk Pegmatite. XXXI. G.F.F. 64, p. 19.

29. Microcline perthite

The potassium feldspars of the Varuträsk pegmatite only occur in perthitic development. They are one of the dominating minerals of the pegmatite, particularly, abundant in the eastern wing, on which they bestow a characteristic feature on account of the huge size of anhedral masses and individual crystals. A single crystal measured 320 cm and was even then only partially exposed. (See Figs. 40 and 42 from the hanging wall of quarry P.)

The prevailing colour of the perthite is light salmon-coloured, more seldom it is almost pure white. The perthitic texture is generally observable with the naked eye.

On account of the constant amount of rubidium in the perthite (mean of 11 analyses 2.2% Rb₂O) it has been signified as rubidium-microcline perthite in accordance with the name "microcline rubidifière" given by Vernadsky for analogous feldspars from the Ilmen mountains in the Ural (V. Vernadsky, 1913). A certain amount of caesium is also present in all the six analysed specimens of the perthite. The higher percentage thereof is always combined with the higher content of rubidium.

Cross twinning of exceptional regularity is often apparent under the microscope.

Adamson has given a detailed description of the perthite, including many chemical and optical determinations (O. J. Adamson, 1942). It would here lead too far to recapitulate his results and conclusions. Reference must in this connection be made to his original paper (i.e.). In the following only some summary data will be referred to.

The extinction angles of the microcline on (001) vary between 18°–20° and on (010) between 3°–6°. The axial angles 2V_α vary between 78°–82°. Similar determinations of the perthitic albite gave extinction angles on (001) between 2° and 5° and on (010) between 16° and 20°. 2V_α varies between 76°–80° (in all cases the result of 11 separate determinations, cf. Adamson, i.e. p. 26).

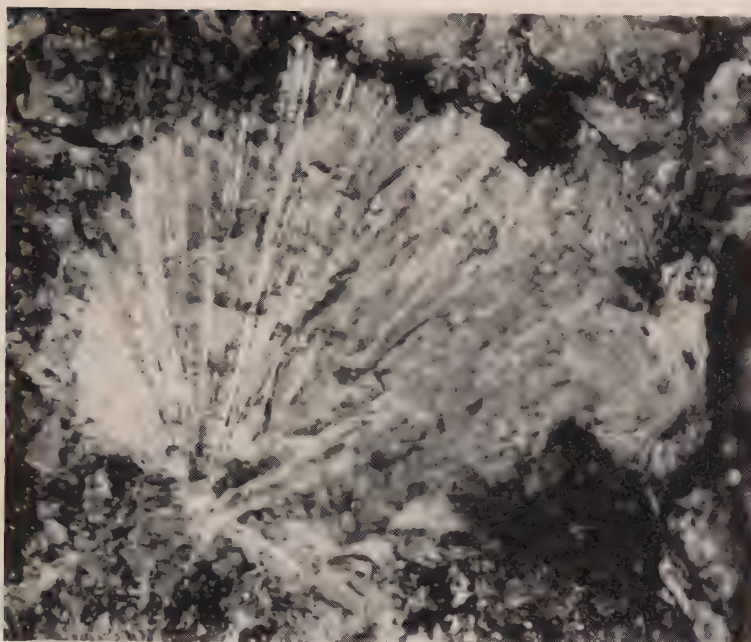


Fig. 39. Spheroidal burst of cleavelandite. Quarry D. $\frac{6}{7}$. Photo S. Hedlund.



Fig. 40. Microcline perthite crystal. Longest extension 320 cm. Northern wall of quarry P.
Photo G. Gustavsson.

Table 32. Analyses of microcline perthite.

	1	2	3	4	5	6
SiO ₂	63.58	64.84	64.02	64.89	63.83	64.07
TiO ₂	0.00	0.00	0.00	0.00	0.00	0.00
ZrO ₂	—	—	—	0.00	0.00	0.00
Al ₂ O ₃	17.80	18.91	19.29	18.68	18.61	18.99
Fe ₂ O ₃	0.03	0.02	0.04	0.12	0.08	0.03
FeO	—	—	—	—	—	—
MnO	0.01	0.00	0.002	0.00	0.00	0.00
CaO	0.40	0.01	0.07	0.15	0.07	0.11
MgO	0.04	0.01	0.01	0.00	0.00	0.00
BaO	—	—	—	0.00	0.00	0.00
Li ₂ O	0.34	0.03	0.06	0.03	0.04	0.05
Na ₂ O	0.50	3.75	1.43	3.72	1.62	1.17
K ₂ O	12.30	11.32	13.95	10.51	13.20	12.57
Rb ₂ O	3.30	0.64	0.68	1.20	1.40	2.10
Cs ₂ O	0.60	0.10	0.10	0.49	0.54	0.60
Tl ₂ O	—	—	—	—	—	0.02
P ₂ O ₅	0.64	0.33	0.25	0.32	0.37	0.25
F	0.03	0.02	0.02	0.01	0.05	0.02
H ₂ O ⁺	0.64	0.13	0.22	0.24	0.15	0.22
H ₂ O ⁻	0.20	0.03	0.09	0.02	0.02	0.22
	100.41	100.14	100.23	100.38	99.98	100.20
Sp. gr.		2.572				2.524

Analyses of microcline perthite, Varuträsk. Thelma Berggren anal. Analyses Nos. 2 and 3 were from the western wing of the pegmatite, all the others from the eastern wing.

Table 33. The alkali and lime content of microcline perthite from Varuträsk.

	Li ₂ O	Na ₂ O	K ₂ O	Rb ₂ O	Cs ₂ O	CaO	Na-f	K-f	Rb-f	Cs-f	Ca-f
1.	0.34	0.50	12.30	3.3	0.6	0.40			11.49	1.56	
2.	0.05	1.17	12.57	2.10	0.00	0.11	10.59	79.32	7.78	1.71	0.60
3.	0.04	1.57	—	1.7	0.7	n.d.	13.26	78.97	5.91	1.86	n.d.
4.	0.05	1.94	—	1.7	0.7	n.d.	16.30	75.93	5.91	1.86	n.d.
5.	0.06	1.59	—	1.5	0.7	n.d.	13.41	79.54	5.19	1.86	n.d.
6.	0.04	1.62	13.20	1.4	0.5	0.07	14.08	79.21	4.95	1.43	0.33
7.	0.03	3.72	10.51	1.2	0.5	0.15	31.54	62.28	4.16	1.27	0.75
8.	0.04	3.40	—	1.19	0.4	<0.01	28.72	66.01	4.15	1.12	<0.05
9.	0.03	2.44	—	1.14	0.7	<0.01	20.60	74.10	3.96	1.34	<0.05
10.	0.04	2.70	—	1.14	0.3	<0.01	22.80	72.42	3.96	0.82	<0.05
11.	0.03	3.11	—	1.00	0.2	<0.01	26.13	69.73	3.44	0.52	<0.05
12.	0.01	3.28	—	0.91	0.2	<0.01	27.72	68.58	3.18	0.52	<0.05
13.	0.06	1.48	—	0.91	0.4	<0.01	12.52	83.18	3.18	1.12	<0.05
14.	0.04	2.90	—	0.86	0.15	<0.01	24.53	72.14	2.96	0.37	<0.05
15.	0.01	3.11	—	0.75	0.2	<0.01	26.31	70.57	2.60	0.52	<0.05
16.	0.06	1.43	13.95	0.68	0.1	0.07	12.44	84.52	2.40	0.30	0.34
17.	0.03	3.24	—	0.65	0.2	<0.01	27.42	69.79	2.27	0.52	<0.05
18.	0.02	2.73	—	0.65	0.2	<0.01	23.06	74.15	2.27	0.52	<0.05
19.	0.03	3.75	11.32	0.64	0.1	<0.01	31.35	66.12	2.17	0.30	0.06
20.	0.01	2.70	—	0.58	0.2	<0.01	22.80	74.67	2.01	0.52	<0.05
21.	0.08	2.97	—	0.58	0.15	<0.01	25.10	72.52	2.01	0.37	<0.05

The analyses are now grouped according to decreasing content of Rb₂O. Analyses Nos. 1–10 and 14 are from specimens from the eastern wing (quarry P). Nos. 11, 16, 19 and 21 are from the western wing (quarry D). Nos. 12, 13, 15, 17, 18 and 20 are from specimens within the wall zone. In the Analyses 3–5, 8–15, 17–18 and 20–21 K₂O values were not given. In these analyses K-f has therefore been calculated, subtracting the sum of Na-f Rb-f and Cs-f from 100.

Nine further partial analyses of specimens from the eastern quarry P gave between 2.4 and 1.16 % Rb_2O . For further details see Adamson (l.c., p. 37).

Adamson observes that the extinction angles on (001) are somewhat higher than the values generally obtained and believes the divergence to be due to their high content of rubidium and caesium. He assumes that it would be surprising if this should not cause some change in the position of the indicatrix.

Miss Berggren has made six analyses of the microcline perthite (Table 32). Beside this Adamson has made no less than 15 new determinations of the alkalies and 9 partial analyses of the rubidium content. Samples were taken from all parts of the pegmatite. The content of Rb_2O varied from 0.58 % to 3.3 % and of Cs_2O from 0.1 to 0.7 %.

Adamson has tabulated the content of the alkalies and lime in these 21 analyses and calculated the weight percentage of the feldspar components, signified as Na-f, K-f, Rb-f, Cs-f and Ca-f. His figures are given in Table 33.

It is of interest to note that with only one exception in the analysed material (No. 21) the higher content of rubidium in the perthite occurs in samples from the eastern wing, which also contain the relatively higher content of caesium.

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 QUENSEL, P. (1937), Minerals of the Varuträsk Pegmatite VIII. G.F.F. 59, p. 455.
 VERNADSKY, V. (1913), Bull. Soc. Fr. de Min. 60, p. 263.

IV. The paragenesis of the Varuträsk pegmatite

1. Terminology referring to phases of mineralisation in zonal pegmatites

Before entering upon the paragenetic description of the Varuträsk pegmatite some references may be advanced on prevailing conceptions regarding the paragenetic development of zonal pegmatites and on the terminology which has been applied to different stages of the same.

A partition into different phases has in general been referred to a temperature scale. This has foremost been elucidated by Fersman. In his well-known paper *Über die geochemisch-genetische Klassifikation der Granitepegmatite*, published in *Min.-petrogr. Mitt.* (Vol. 41, 1931), he has expounded his conception regarding the geochemical differentiation of granite pegmatites as well as the temperatures which he assumes to have been decisive for the formation of certain minerals or mineral assemblages. This paper was a summary by Fersman of his contemporaneous extensive monograph published in 1931 in Russian. In 1951 a complete translation of this work was published in French by R. du Trier de Terdonck and J. Thoreau under the title *A. E. Fersman, Les pegmatites, leur importance scientifique et pratique* (Rapid copy, Louvain 1951. 675 pages, 101 diagrams and 32 tables). References in this paper regarding Fersman's comments refer to this translation.

The inversion point of quartz (around 600°) and the critical temperature of water (around 400°) are by Fersman taken to represent the main points of division in the paragenetic evolution of granitic pegmatites. The first signified the partition between his epimagmatic and pneumatolytic stages. The second separates the pneumatolytic stage from Fersman's hydrothermal stage. Regarding the latter he says: "Le point critique se trouve à 374° avec une pression de vapeur saturée de 219 Atm. L'accroissement de la pression la port jusqu'à 400°-425° et l'intervention de matières en solution peut la porter encore plus haut" (p. 65).

The schema below exemplifies the principle features of Fersman's classification

	Epimagmatique		Pneumatolytique			Hydrothermale		
	800°	700°	600°	500°	400°	300°	200°	100°
Phases :	B	C	D-E	F-G	H	I	K	
	épimag-	pegma-	pegma-	hyper-	haut	moyenne	basse	
	matique	titique	toide	critique	température			
	propre	propre						

Regarding the partition between the stages "pneumatolytique" and "hydrothermale" he remarks: "La limite entre G et H est marquée par une discontinuité nette dans la majorité des processus pegmatitiques qui ne reviennent plus à leur formes antérieure, mais correspondent sans doute à un état autre, se marquant par toute une série de caractères morphologiques et géochimiques spéciaux dont l'analyse m'oblige à relier ce point à la température critique de la vapeur d'eau" (p. 59).

Partitions of similar nature are obvious in the paragenetic development of the

Varuträsk pegmatite. The partition between Fersman's pegmatitic and pegmatoid phases C and D coincides with the division between what in this paper is signified as the pegmatitic and the pneumatogenic stage, exemplified by an obvious break in the sequence of mineralisation and an exchange of participating mineral assemblages. Likewise Fersman's accentuated partition between his "hypercritique" and "hydrothermale" phases G and H corresponds in every detail with another marked breach in mineralisation in the Varuträsk pegmatite, manifested by the deposition of many new minerals.

Regarding the terminology of the different stages in the evolution of granite pegmatites Fersman definitely restricts the term hydrothermal to such stages of mineralisation which have taken place below the critical point of aqueous solutions. He remarks thereon: "Une contestation a surgi en ces derniers temps autour du terme hydrothermale, que Larsen et aussi Lindgren et Hess-Ros sont proposé d'appliquer également au-dessus du point critique d'ébullition, de ce qui les amène à parles de processus hydrothermeaux de haut temperature au lieu de pneumatologiques. Schneiderhöhn a tout à fait raison lorsque il s'insurge contre l'altération inutile des termes (p. 66)."

The term hydrothermal has also in many more recent publications been extended to phases of mineralisation pertaining to temperatures above the critical temperature. It is evident that when Landes says that after "a first class, solidified as magma ... the next three classes represent successive waves of hydrothermal activity from a slowly cooling igneous mass beneath" (31, p. 356), the term hydrothermal is used in a very different sense from that applied by Fersman. Heinrich evidently uses the term in the same meaning when in a table of the paragenesis of the minerals of Eight Mile Park, Colorado, he denotes that a first hydrothermal phase directly follows the magmatic stage (20, p. 584). Shand follows the same terminology when saying that a first and second magmatic stage are followed by a deuteric or high temperature hydrothermal stage and a low temperature hydrothermal stage (64, p. 350).

Shand says in the same connection that one "would gladly follow Fersman if his phases could be separated as easily in practice as on the paper". On the other hand Turner-Verhooogen say: "the most satisfactory scheme (as to the possible sequence of crystallisation in pegmatites) is that of Fersman" (Igneous and metamorphic Petrology, 71, p. 331).

Further quotations might be offered but those given may suffice to exemplify the differences in terminology used by different authors regarding certain phases of replacement in pegmatites.

Contrarily to Shand's distrust regarding the practical application of Fersman's phases, the sequence of mineralisation at Varuträsk so manifestly corresponds to his phases that coincidences can be followed even in many details.

In this paper the significance of the term hydrothermal will, in accordance with Fersman, Schneiderhöhn and others, be restricted to denote the replacements in the pegmatite which have taken place below the critical temperature of the mineralising aqueous solutions instead of being extended to include "high temperature hydrothermal stages", following the magmatic stage. Such a stage will instead be assigned to processes of pneumatolytic activity.

The terms pneumatolytic and hydrothermal will however not be applied in this paper to signify limited stages of mineral deposition in the Varuträsk pegmatite.

The term pneumatolytic has so universally been applied in a much wider sense in connection with different geological processes that it now hardly can be taken as expressive for a definite phase of mineralisation in a pegmatite. Instead the term *pneumatogenic* will be applied to the phase of mineral deposition which has taken place during the interval between around the inversion point of quartz and the critical temperature of the aqueous solutions, i.e., between around 600° and 400°. The name is meant to indicate that the replacements of this stage pertain to the general conditions prevailing during pneumatolytic activity.

Conformably, the term *hydatogenic* will be used to signify the phase of mineralisation which has taken place below the critical temperature of the aqueous solutions, i.e. between 400° and 100°. The temperature intervals are however assumed only to represent approximate values. Too many uncontrollable influences of pressure, concentration and equilibrium may shift any fixed temperature scale in one direction or another.

The paragenetic classification of the Varuträsk pegmatite will have the following aspect:

The names zones and units will be used in the meaning proposed by Shainin and Heinrich. Zones refer to the different phases of zonal development during the primary consolidation of the pegmatite whereas units signify different phases of succeeding replacements.

The zonal development of the pegmatite will be named the *pegmatitic stage*. The gradual differentiation and fractional crystallisation of this stage is assumed to have taken place under epimagmatic conditions, i.e. above around 600°.

The following stage will be named the *pneumatogenic stage* as accounted for above. This stage will include all replacement units which can be assumed to have taken place at temperatures below those ascribed to the pegmatitic stage but above the critical temperature of the mineralising aqueous solutions. This stage has been subdivided into a high temperature phase and a lower temperature phase, between which a definite break in mineralisation is obvious, as will be elucidated in the following description.

Subsequent alterations due to the activity of thermal water of hypogene origin will be attributed to a *hydatogenic stage*.

A final stage due to the action of percolating ground water or to superficial weathering will be signified as the *stage of supergene alterations*.

As the individual minerals in the pegmatite are summarily described in the preceding part of this paper, further comments thereon will in the following be restricted to such circumstances as have import with regard to the paragenetic development of the pegmatite.

2. The pegmatitic stage

The pegmatitic stage is taken to represent the initial zonal consolidation of the pegmatite in a closed system. An increasing content of volatiles in subsequent residual fluids, containing additional compounds, are assumed to be responsible for the different mineral assemblages in the different zones. The border and wall zones, the intermediate zones and the core denote the sequence of consolidation from the wall inwards.

amphibolite
(surrounding wall)

Border zone

Wall zone



Fig. 41. Contact between amphibolite and wall zone. Between these a narrow fine-grained border zone is observable. Nat. size. Photo O. Mellis.

a. The border zone

The border zone of the Varuträsk pegmatite can be traced nearly everywhere where the contact against the surrounding amphibolite is exposed. The characteristic feature of this zone is the very fine-grained development of its mineral assemblage, due to the cooling influence of the surrounding rock formation. The border zone seldom attains more than some 10 cm in thickness, often it is less than one cm thick (Fig. 41).

The mineral assemblage is simple and uniform. The only essential constituents of primary origin are quartz and muscovite. Accessory minerals are all but absent.

b. The wall zone

In one sense one may say that the border zone and the wall zone co-ordinate inasmuch as the bulk mineral composition is the same, though in the wall zone developed in larger individuals. This on the other hand establishes a sharp boundary between the two zones.

The thickness of the wall zone may vary from some 5 dm up to several meters. An intervening part between the eastern and western expansions of the pegmatite consists only of the wall zone mineral assemblage, which there can attain a thickness of over five meters.

In many places the wall zone consists exclusively of quartz and muscovite. The muscovite then occurs in large silvery white books, up to one dm in width.

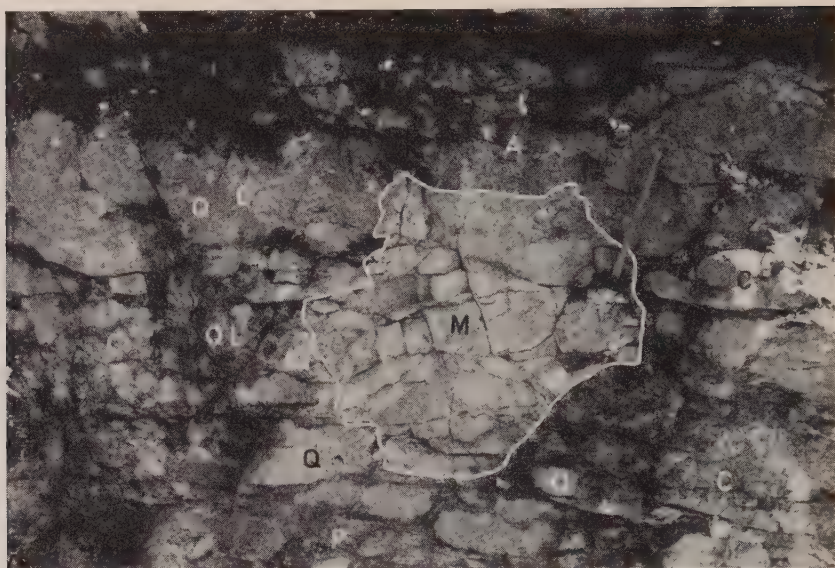


Fig. 42. Microcline perthite merging into the quartz-muscovite assemblage of the wall zone. Quarry around D.

A, wall zone; M, microcline perthite crystal (diameter 90 cm); Q, quartz. (Later replacements: P, petalite; C, cleavelandite; QL, quartz-lepidolite.) $\times 1:20$. Photo G. Gustavsson.

Other minerals, however, now locally appear. The most usual and at the same time the most characteristic of them is black tourmaline, which in its distribution in the Varuträsk pegmatite is principally restricted to the wall zone. Where it occurs in larger quantities, it gives the mineral assemblage of this zone one of its characteristic features. Near the border zone the black tourmalines often show a tendency to attain a longitudinal orientation, vertical to the contact. (See Fig. 41.)

Locally beryl is also found partaking in the primary mineral assemblage of the wall zone. The most notable occurrence is around excavation H_1 , where the mineral is developed in large crystals, up to several dm in length, contrasting both in form and size with the beryls pertaining to later stages of development.

A question worth notice is the occurrence of large crystals of microcline perthite amongst the mineral assemblage of the wall zone. Adamson has in his paper on the feldspar group of the Varuträsk pegmatite a photograph (Fig. 42) showing a large microcline perthite crystal merging into the quartz-muscovite assemblage of this zone, close to the hanging wall. Minerals of later replacement units intergrade, but as they belong to later stages in the development of the pegmatite, they are without significance in the present connection. The problem in question is if microcline perthite, as seen in the photograph and often observed in the field, belongs to the original mineral assemblage of the wall zone or if its mis-en-place there is the result of later replacement.

Mostly on account of microcline perthite being only locally present among the mineral assemblage of the wall zone, and then chiefly along the hanging wall, whereas in other places it is conspicuously absent, it could reasonably be postulated that the microcline has been intercalated by metasomatic replacement through residual solu-



Fig. 43. Northern wall of quarry P showing huge crystals of microcline perthite together with large amounts of quartz. The microcline perthite crystal to the left is the same as pictured in Fig. 40. Photo G. Gustavsson.

tions emanating from the adjacent intermediate zone. This supposition would naturally only refer to isolated individuals occurring within the typical quartz-muscovite assemblage of the wall zone. Where this zone grades inwards into the intermediate zone, an interspersing of mineral constituents is to be expected and is often observed.

A further circumstance which may be taken to indicate that the isolated crystals of microcline perthite in the wall zone are due to replacements is that the crystals there attain dimensions extraneous of the wall zone in general but characteristic of the sequent intermediate zone.

A circumstance worth attention is the chemical composition of muscovite within the wall zone. An analysis of this mica from near the foot wall of the eastern wing shows a not insignificant content of lithium (0.73 % Li_2O) indicating that already during this initial stage of consolidation the participating fluids contained a certain amount of lithium.

c. The intermediate zones

The intermediate zone includes the zonal development, intermediate between the wall zone and the core. At Varuträsk, as is the case in most other complex pegmatites, it forms the greater mass of the pegmatite.

A sub-division into two phases can be discerned, denoted as an outer and an inner intermediate zone. The difference between them is that the outer zone has a simpler mineral composition than the inner zone.

The essential minerals of the outer zone consist predominantly of microcline perthite, quartz and muscovite. The difference between the wall zone and the outer intermediate zone is that microcline perthite now enters as the dominant mineral, developed in anhedral masses or crystals of great size. A single crystal seen in Fig. 43 measures over 3 meters in length and is then only partly exposed.

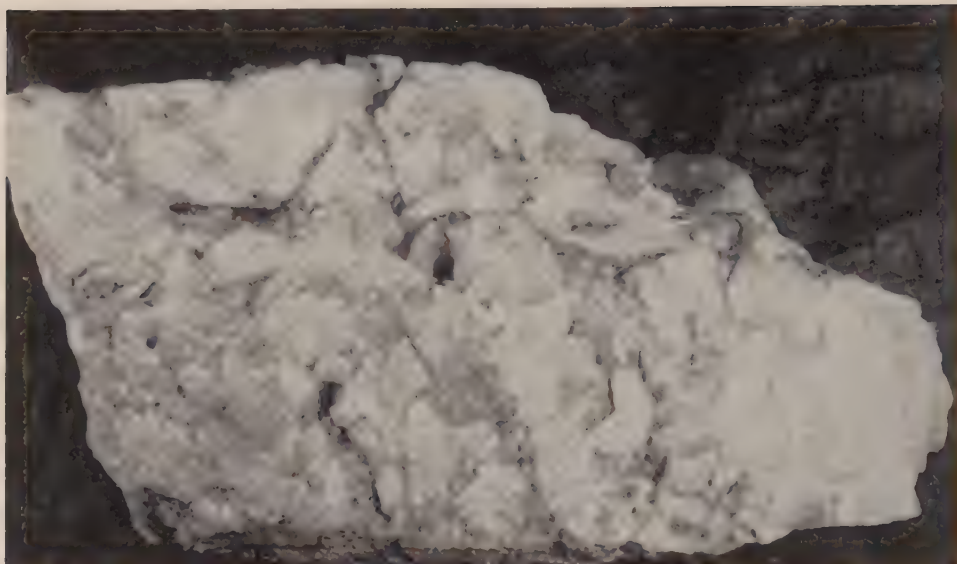


Fig. 44. Older generation of spodumene in thick tabular masses, amply interspersed with quartz.
Nat. size. Photo O. Mellis.



Fig. 45. Anhedral masses of montebrasite belonging to the inner intermediate zone of the pegmatitic stage. Longest extension of exposed part 130 cm. Northern wall of quarry P.
Photo G. Gustavsson.

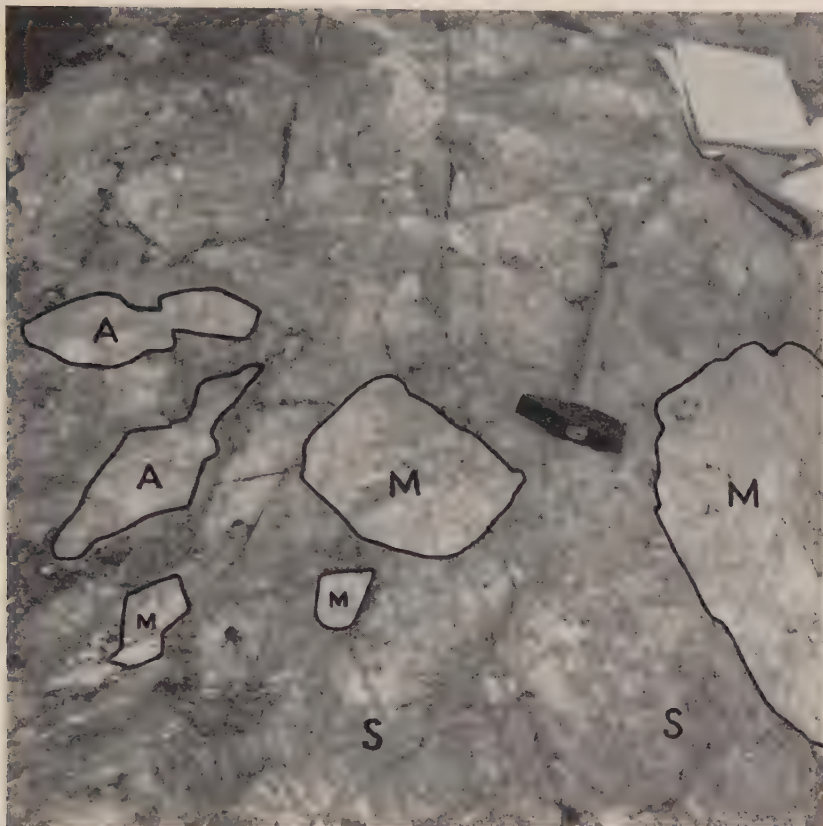


Fig. 46. Cleaned rock exposure 30 m NE of the shaft in the eastern wing. Corroded fragments of montebrasite (A) and angular pieces of microcline perthite (M), encompassed in younger spodumene (S) replacing the mineral assemblages of the intermediate zones. $\times 1/20$. Photo: B. Tornqvist.

The mineral assemblage of the inner zone is the same as that of the outer zone with the addition of some pronounced lithium-bearing minerals, evidently due to an additional content of lithium in the residual solutions. The essential minerals in this respect are spodumene and montebrasite, both present in considerable amounts.

In other respects the outer and inner intermediate zones show no dissimilarities. A characteristic feature for both zones is the huge size of most of the participating minerals, presumably due to a concentration of volatiles in the residual solutions of the pegmatitic stage, combined with a subsequent slower rate of consolidation.

Attention may be called to the considerable amount of rubidium present in the microcline perthite as well as to minor quantities in the mica minerals of this stage. The content of this alkali, combined with a certain amount of caesium is obviously greater in the perthite in the eastern part of the pegmatite than in the western part. The medium of nine analyses from the eastern wing gave 1.55% Rb_2O (maximum 3.3%) whereas the medium of four analyses of corresponding material from the western wing was only 0.72% (maximum 1.0%; cf. Adamson 1942, p. 37).



Fig. 47. Fragments of microcline perthite in spodumene. Nat. size. Photo O. Mellis.

d. The core

The core of a pegmatite generally represents the ultimate stage of consolidation of a zonal pegmatite. In most pegmatites where this zone is developed, the mineral assemblage thereof has a tendency towards simplification in composition and to become more siliceous. The core may then often consist of only quartz. This is the case in the Varuträsk pegmatite where the core is almost exclusively composed of pure milky quartz.

As recorded from many other flat-lying or moderately dipping zonal pegmatites, the core at Varuträsk is not centrally located but displaced towards the southern foot wall of the eastern wing where it occupies a lens-formed body about 50 m in length and 15 m in breadth. During mining processes around the large quarry P, many thousand tons of core quartz have been excavated.

Though surrounded by the mineral assemblages of later replacement units and locally intersected by minerals of the same, the core on the whole shows but insignificant signs of replacement by invading solutions of succeeding phases of mineralisation.

e. Sequence of mineralisation

Before entering upon some details regarding the sequence of mineral deposition during the pegmatitic stage, a few observations may be advanced on some significant minerals pertaining to this stage.

In the first place spodumene may be taken into consideration. As already noted in the mineral description this mineral occurs in two different modifications, of which

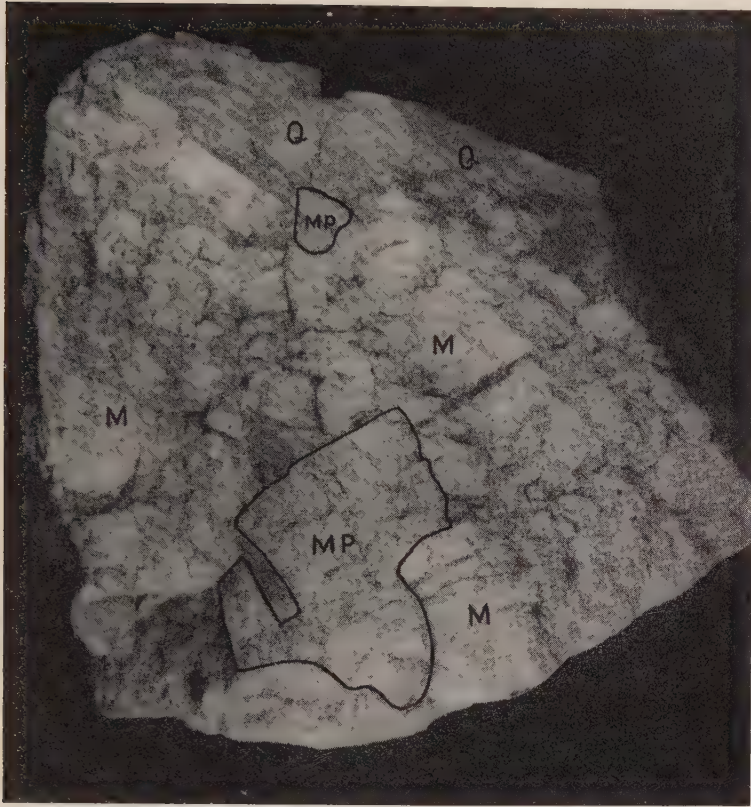


Fig. 48. Fragments of microcline perthite (MP) encompassed in massive montebrasite (M), subsequently replaced by vitreous quartz (Q). $\times \frac{3}{4}$. Photo O. Mellis.

one is a characteristic constituent of the inner intermediate zone whereas the other pertains to a subsequent replacement unit. The type which occurs in the pegmatitic stage is always developed in thick tabular masses, amply intermingled with quartz, often in such amounts that the two constituents may occur in nearly equal proportions (Fig. 44). This development is probably very similar to what Stockwell has described from Bear Lake Mineral Claim in Manitoba under the name of "quartz-spodumene" (68, p. 30). When recurring in the pneumatogenic stage the spodumene shows a very different aspect, then developed in slender semi-transparent laths (see Fig. 53).

Montebrasite likewise occurs in two generations of somewhat different aspect. When together with spodumene, pertaining to the inner intermediate zone of the pegmatitic stage, it occurs in anhedral masses (Fig. 45) or in large coarse crystals, milky white in colour and with a greasy lustre. In a younger generation, it instead occurs in small crystals, purer white in colour and with a more vitreous lustre.

The third mineral worth observation in this connection is the microcline perthite which occurs throughout the combined intermediate zones in equivalent amounts. A certain difference may however be observed in that the perthite in the outer

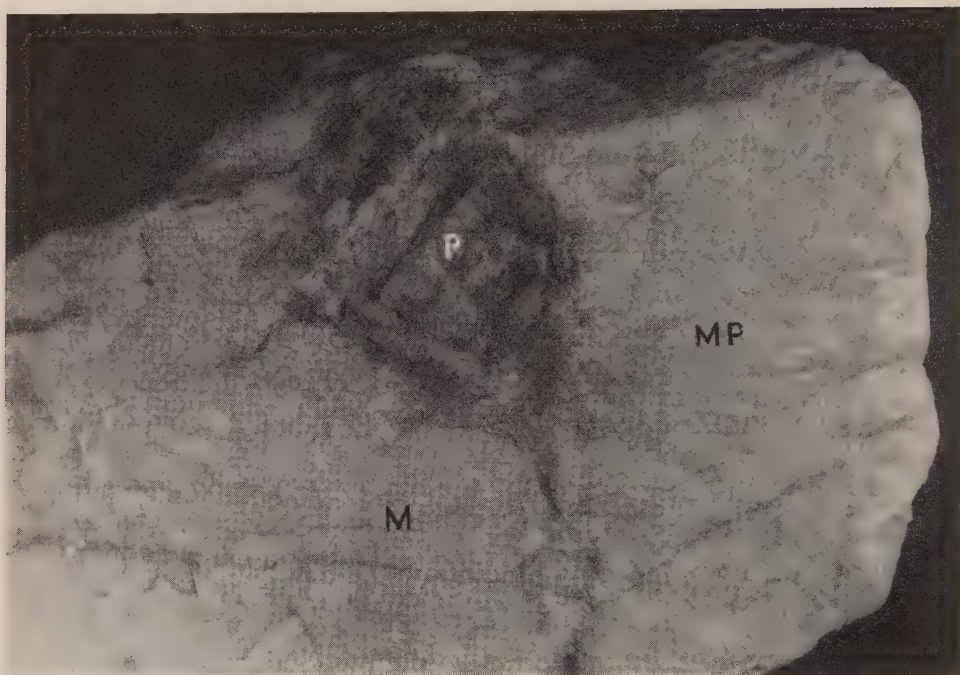


Fig. 49. Massive montebrasite (M) in contact with white microcline perthite (MP). A nodule of petalite (P) is here incidental, belonging to a subsequent replacement unit. Quarry P. $\times \frac{4}{5}$
Photo O. Mellis.

zone has a characteristic light salmon colour whereas in the inner zone it is, at least in general, purer white. This difference may perhaps be attributed to variable amounts of the rare alkalis in the solutions during the consolidation of the pegmatitic stage.

In the following pages some photographs illustrating the sequence of crystallisation during the pegmatitic stage are reproduced.

In 1951 the Boliden Company laid bare a small rock area, clean-faced by glacial erosion, situated some 30 m NE of the shaft in the eastern wing. Corroded fragments of montebrasite as well as more angular pieces of microcline perthite are there seen encompassed in a large mass of spodumene. (Fig. 46.) As the spodumene here has the habit, characteristic for its apertenance to the pneumatogenic stage, the enclosed fragments must belong to a preceding stage of mineralisation. According to what has been stated above the montebrasite would pertain to the inner intermediate zone whereas the perthite, on account of its colouring may be assumed to have originated in the outer zone as would also be the case in a corresponding specimen (Fig. 47) from near the same locality.

Fig. 48 shows fragments of microcline perthite enclosed in massive montebrasite. In this case the perthite has again the light salmon colour characteristic of its occurrence in the outer intermediate zone whereas the montebrasite has the typical development in the inner intermediate zone. The perthite can here be clearly distinguished megascopically from the montebrasite by its colouring.



Fig. 50. Montebrasite brecciated by vitreous quartz. Nat. size. Photo O. Mellis.

This is on the other hand not the case in the specimen pictured in Fig. 49, where pure white microcline perthite is seen in contact with montebrasite of the same nature as in Fig. 47. In this case the colouring of the two minerals is so much the same that they are at first sight hardly distinguishable megascopically from one another. In accordance with the assumption advanced above, this would imply that both minerals belong to the inner intermediate zone. The petalite in the photograph is incidental since pertaining to a subsequent stage of mineralisation.

Fig. 50 shows montebrasite of the pegmatitic stage brecciated by gray vitreous quartz. No obvious replacement has taken place as one can rejoin many of the isolated pieces. Around the quarry P such specimens were during mining of frequent occurrence. They much resemble a specimen from Peerless Dike, Keystone, described by Hess, though in that case amblygonite is replaced by albite (23, Fig. 2).

Summarizing the most characteristic features of the pegmatitic stage of mineralisation the following instances may be set forth:

The mineral composition and sequence of mineralisation of the border and wall zones and of the core present no complicated features and need not further be commented on. The points of interest with regard to the paragenesis are concentrated to the intermediate zones.

Microcline perthite is the predominant mineral of both the outer and the inner intermediate zones, developed in huge individuals associated with high temperature quartz and mica minerals. The latter can not be classified as lepidolites but must be referred to as muscovites with an insignificant content of lithium, as was already the case with the muscovites of the wall zone.

In the inner intermediate zone a first generation of spodumene and montebrasite occur indicating an increase of lithium in the residual solutions. Montebrasite can in this zone attain very large dimensions. Individual crystals may measure over 50 cm in diameter.

An interspersation of mineral components between the inner and outer intermediate zones is often to be observed, due to the circumstance that before the ultimate consolidation of the inner zone residual solutions have invaded and replaced already consolidated minerals of the outer zone in accordance with what previously has been anticipated regarding the occurrence of microcline perthite amongst the mineral assemblage of the wall zone. Circumstances of this nature often cause difficulties in separating the zones.

The primary fractional differentiation of the pegmatitic stage as well as the secondary reactions between its mineral assemblages are assumed to have taken place under conditions prevailing in a closed system. No indications have been observed which might suggest that additional material has been added during the sequence of mineralisation of the pegmatitic stage. The local reactions can in every instance be attributed to an interchange of already existing material within a closed system.

3. The pneumatogenic stage

a. General features

In accordance with the scheme given above, the term pneumatogenic stage will here be used to denote the phases of replacement which follow the zonal consolidation of the pegmatitic stage. A definite break in the mineralisation has, according to all field observations, occurred after the completion of the zonal development, which was in all phases consolidated before the renewed activity of the pneumatogenic stage set in.

Whereas the temperatures prevailing during the pegmatitic stage are taken to have exceeded the 600° limit, the replacement units of the pneumatogenic stage are postulated to have taken place between this limit, and the critical temperatures of the co-operating solutions, i.e. between approximately 600° and 400°.

Another essential difference between the development of the two stages is obvious. Whereas the consolidation during the zonal development of the pegmatitic stage represents a continuous process of mineralisation during slowly progressing differentiation in closed system conditions, the renewed activity of the pneumatogenic stage displays successive depositions, replacing pre-existing mineral assemblages and depositing new minerals in their stead. The phases of such replacements are signified as replacement units.

A factor in trying to establish the sequence of the different replacement units has been the supposition that a gradual decrease in temperature occurs during this stage. A bipartition has therefore been postulated between a high and a lower temperature phase. The mineralisation during the high temperature phase has in general taken the form of extensive bulk replacements, whereas the mineral deposition of lower temperature phases is more restricted to fracture-controlled replacements.

According to field indications a further partition of the high temperature phase into two units seems justified. During a first unit the solutions have in respect of their content of alkali mainly carried lithium. During a somewhat later invasion with a more restricted distribution the solutions have preferentially contained caesium. The two units have been named the lithium replacement unit and the caesium replacement unit. The names, however, must not be taken to imply that these alkali components are not present in other phases, but to accentuate that these units are characterised by containing the concentrated amounts of these alkalies.

The dominant lithium-bearing minerals of the lithium replacement unit are lepidolite, petalite and a second generation of spodumene, whereas pollucite is the dominant mineral of the subsequent caesium replacement unit.

The prevailing and most characteristic mineral of the low temperature replacement unit is albite, mainly developed in the form of cleavelandite. This phase has been named the sodium replacement unit, representing the first concentrated sodic influx during the paragenetic evolution of the pegmatite. Mineralisation during this unit has been very extensive and includes a number of minerals, not represented in previous phases. As the main replacements of this unit are concentrated to the western expansion of the pegmatite, many of the rare minerals of this unit are mostly or only found there.

Fluorite is of rare occurrence in the Varuträsk pegmatite. It has only been found in one instance, filling out two small vugs together with black quartz. As vugs are practically non-existent in the pegmatite, the isolated occurrence in question can only signify a trivial phase of mineralisation, representing a concluding phase of mineral deposition during the pneumatogenic stage.

The summary review of the different phases of the pneumatogenic stage now given refers to the general features of its paragenetic development. Under the following headings some detailed observations will be given regarding the characteristic features of the different replacement units.

b. The high temperature phase

1. The lithium replacement unit

The lithium replacement unit is taken to represent the first epoch of renewed mineralisation after the conclusion of the pegmatitic stage which in all details fashioned the zonal development of the pegmatite. The name is, however, only meant to indicate that the main concentration of lithium occurs in this unit, verified by the abundance of the lithium-bearing minerals lepidolite, petalite and a second generation of spodumene, together constituting a characteristic mineral assemblage of this unit.

The greatest concentration of lepidolite occurs in the form of a mauve-coloured, fine-grained, massive lepidolitic rock in the western wing (quarry H₂). Up to this

date many hundred tons of this rock have been mined, awaiting further demand for lithium in industrial processes. The lepidolitic rock of this occurrence is in parts speckled with small grains of cassiterite as seen in Fig. 51. Where this occurs the rock is generally also invaded and replaced by cleavelandite as seen in Fig. 52. As cassiterite is in other places nearly always associated with cleavelandite, pertaining to the lower temperature phase of this stage, it is probable that so is also the case when occurring in the lepidolitic rock in question.

Inclusions of beryl locally occur in some abundance in the same rock. The mineral has here a very different habit than in other occurrences within the pegmatite. It is pure white with a vitreous lustre and a granulated texture, presumably caused, by recrystallisation within the large mass of lepidolitic rock in which the beryl has been engulfed. In such a case this beryl would pertain to the pegmatitic stage and represent casual fragments of the large crystals found in the immediate vicinity.

It seems as if the lepidolitic micas of this unit occur predominantly in the form of this fine-grained rock. The rich assortment of other types of lepidolite and lepidolitic micas in the pegmatite are in general connected with subsequent replacement units.

Together with lepidolite, petalite is the most abundant lithiumbearing mineral of this unit. Though in general of rare occurrence in other lithium pegmatites, it is present in great quantities at Varuträsk. Up to this date some thousand of tons of the mineral have been mined in the northern off-shoot of the eastern wing (quarry D), where it occurs in greatest quantities. As was the case with the lepidolitic rock, the petalite is everywhere traversed by veins or blotched by scattered replacements of cleavelandite, indicating that petalite also definitely belongs to an elder stage of mineralisation than that of the sodium unit.

The third lithium silicate of quantitative importance pertaining to this unit is spodumene. This mineral has already been found to occur in the inner intermediate zone of the pegmatitic stage. When now recurring in this unit, it is developed in an obviously different habit. Instead of the compact tabular masses of the pegmatitic stage, it now occurs in the form of slender laths (Fig. 53), which, when uncontaminated, are semi-translucent, in contrast to the elder generation of the mineral. It is, however, not unusual that the spodumene of this unit is highly altered to a mixture of clay minerals (rotten spodumene). This is never found to be the case with spodumene of the pegmatitic stage (cf. Fig. 29, p. 65).

No significant indications of a sequence in deposition between spodumene of this unit and petalite have been observed. In most cases the two minerals occur in disconnected areas. Where they occur together they are nearly always found separated by veins of quartz. At one locality, however, they occur intercalated. The right-hand part of Fig. 2, Plate V, is seen partially to consist of a fine-grained intermixture of petalite, spodumene and quartz. The development indicates a contemporaneous deposition. This confirms the supposition that the younger generation of spodumene and petalite pertain to the same replacement unit.

Occasionally petalite has also been observed together with the elder generation of spodumene, pertaining to the inner intermediate zone of the pegmatitic stage as seen in Fig. 54. Under the microscope one may then observe that petalite has along the contact between the two minerals given rise to a narrow zone of reaction within which the spodumene has recrystallised in the form of fibrous sheaves, vertical to the contact. This corroborates that the petalite pertains to a younger phase of mineralisation than that of the pegmatitic stage.

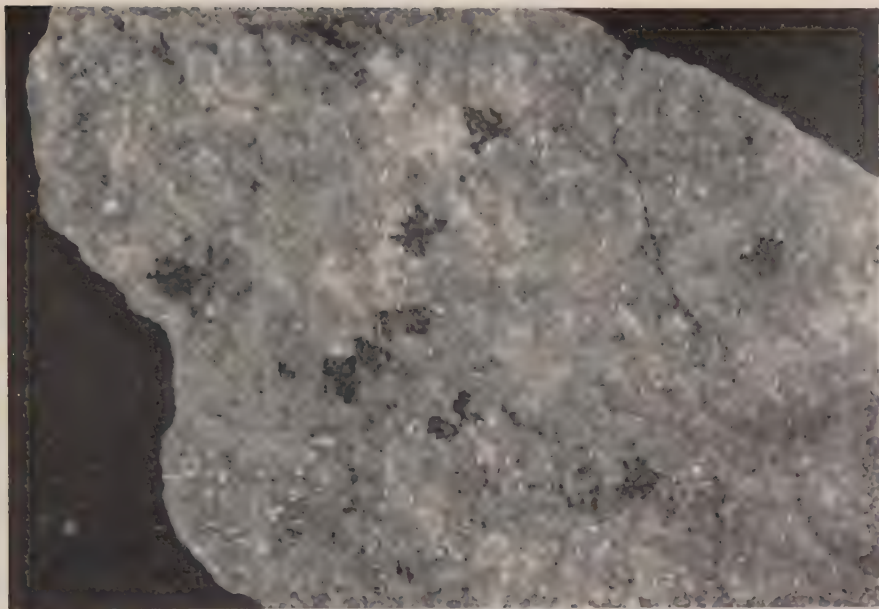


Fig. 51. Fine-grained lepidolitic rock with grains of cassiterite. Quarry H₂. Nat. size.
Photo O. Mellis.



Fig. 52. Lepidolitic rock (gray) replaced by cleavelandite (white). The lepidolitic rock is the same as pictured in Fig. 51. Photo G. Gustavsson.

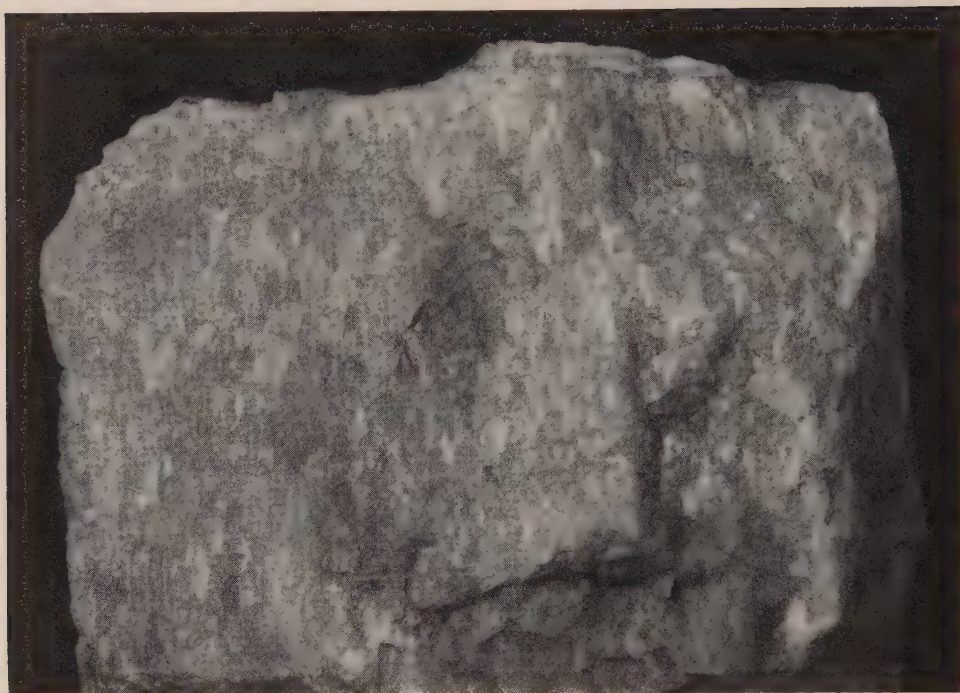


Fig. 53. Younger generation of spodumene. Around quarry P. Nat. size. Photo O. Mellis.

Other minerals pertaining to this unit only occur in trivial quantities. The occurrence of montebbrasite in the inner intermediate zone of the pegmatitic stage has been noted above. A younger generation of the mineral recurs in much smaller crystals in a different habit and lustre. The associated mineral assemblage indicates that the deposition of this type of montebbrasite has preceded the cleavelandite invasion and should therefore pertain to the lithium replacement unit. This type is however of rare occurrence and without significance for the characteristic mineral assemblage of this unit.

Much the same conditions occur regarding beryl. In contrast to the large yellowish crystals of the pegmatitic stage, the mineral now recurs in the form of small semi-transparent individuals. In Fig. 55 a vitreous crystal of this habit is seen enclosed in spodumene, likewise pertaining to the lithium replacement unit.

Of the coloured tourmalines only the green verdelite occurs in this connection. It is, however, not a characteristic species of this unit as it recurs in the lower temperature phase together with rubellite and indicolite as well as there forming the outer shell of the zoned tourmalines.

Manganapatite is of common occurrence, though recurring in much the same habit in the subsequent phase. Manganvoelckerite on the other hand is of rarer occurrence and seems to be restricted to this unit. In cases where the two phosphates are found united, manganapatite always forms the kernel, surrounded by an outer zone of voelckerite denoting a casual deficiency of fluorine during the ultimate period of crystallisation (cf. p. 56).

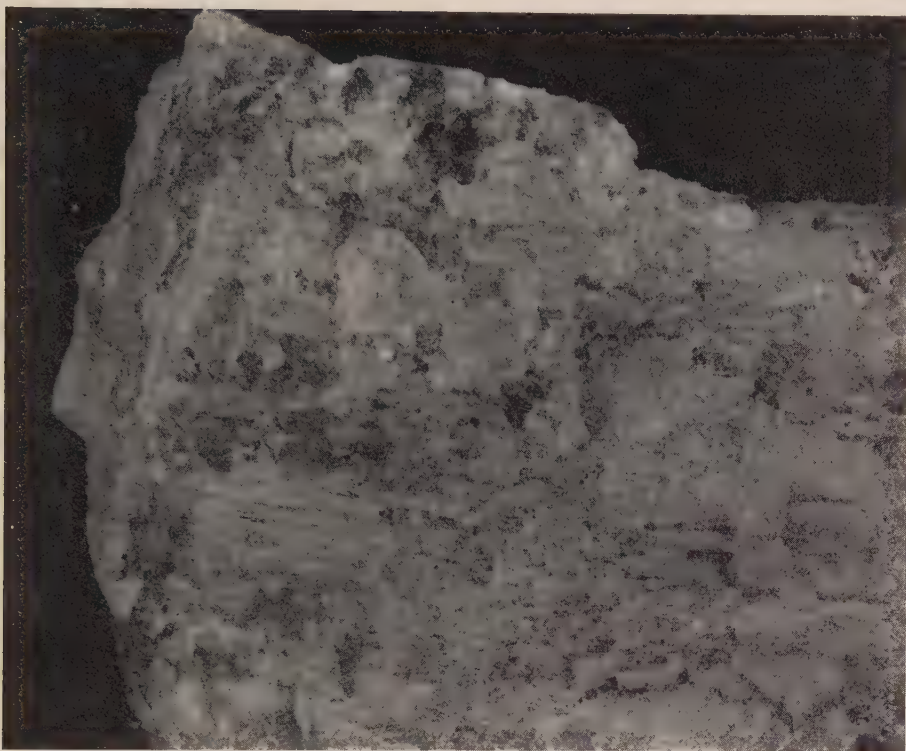


Fig. 54. Intergrowth of petalite (dark) and spodumene. Eastern part of quarry P. Nat. size.
Photo O. Mellis.

The paragenesis of the phosphates triphylite and varulite in the Varuträsk pegmatite offer some problems of special interest.

Triphylite has only been found in some few larger specimens. One of them is pictured in Fig. 20 (p. 40). It is there seen to occur within the typical mineral assemblage of the wall zone. Another specimen, including the largest find of triphylite from Varuträsk (10×15 cm across), shows the same affinity in a still more expressive form. The only other minerals surrounding the triphylite are large books of muscovite and quartz, both in the typical development of the wall zone. Both these finds as well as some few smaller finds of triphylite are all located to a restricted area around quarry H₂. One of the large specimens was found within the wall zone, near the foot wall SW of the quarry, the other within the wall zone near the hanging wall limiting the same quarry to the north.

It is a remarkable incidence that the only occurrences of the rare phosphate varulite coincide with those of triphylite. This can hardly be an accidental circumstance. The problem regarding the formation of varulite has, as noted in the mineral description (p. 47), been clarified through recent investigations by Mason. In accordance with his conclusions varulite must be conceived as an alteration product of lithiophilite which has lately been observed to occur as unaltered kernels in many thin sections of varulite. The size of the varulite pseudomorphs indicates that in



Fig. 55. Vitreous beryl in spodumene. Both minerals pertaining to the pneumatogenic stage. Near point S. Nat. size. Photo O. Mellis.

this case lithiophilite must have occurred in larger individuals and in a different habit than when in a later generation recurring in the form of small nodules in cleavelandite, pertaining to the lower temperature phase of this stage (see Fig. 64). The two modifications are, however, in other respects identical as the diffraction patterns from the different phases coincide.

A significant difference is, however, to be observed regarding the alteration products of lithiophilite in the two occurrences. When associated with triphylite no other signs of alteration can in a first stage be seen than the almost complete change into varulite or subsequently into its oxidation products, alluaudite and purpurite (cf. Fig. 23). When recurring in the later generation, no indications of such alterations are to be seen. Instead the lithiophilite has there succumbed to an oxidation process, resulting in the formation of the minerals sicklerite and purpurite in full accordance with the oxidation of triphylite to ferri-sicklerite and heterosite under the same conditions. A plausible explanation seems to be that the alteration of lithiophilite to varulite has taken place at deeper levels, not subjected to superficial alteration. If subsequently remaining lithiophilite and varulite through erosion have become exposed to levels within the sphere of oxidation, they are found altered to alluaudite and purpurite in full conformity with the oxidation of adjacent triphylite (cf. scheme p. 51).

It is of interest to note that much the same circumstances have been described by Volborth from the Viitaniemi pegmatite in Finland (73, p. 41). He states that in one generation of lithiophilite it is only found altered to sicklerite and purpurite (together with vivianite and strengite). In that case the occurrence is always located to surface exposures. Another generation of lithiophilite was found altered to varulite

along cleavage planes although also showing traces of an alteration to sicklerite. This would in accordance with the supposition given above indicate that first an incipient alteration to varulite has taken place but has been interrupted, and at a later stage the rock assemblage has been exposed to superficial oxidation. Under such conditions it is without importance that the alteration of lithiophilite to varulite has not taken place in the same paragenetic sequence at Viitaniemi and at Varuträsk. Volborth ascribes it to a second generation of lithiophilite, whereas at Varuträsk it occurs in a first generation of the same mineral.

On account of the localised distribution of triphylite and varulite, in main bordering the massive lepidolitic rock around quarry H₂, one might assume that the formation of these minerals has in some way been connected with the deposition of this bulk replacement of lepidolite. They are, however, not found within the lepidolitic rock itself but in places extended outside its contact with the adjacent wall zone. It might be plausible to assume that residual solutions, connected with the deposition of the large masses of lepidolitic rock, can have disseminated in outward direction and there deposited the phosphate minerals.

It is, however, in that case not easy to explain how the alteration of lithiophilite to varulite has taken place, in which Li₂O has been substituted by (Na₂O, CaO). This chemical change can hardly be connected with residual solutions emanating from the deposition of the lepidolitic rock masses. The alteration of a primary deposited lithiophilite to varulite must presumably be attributed to a subsequent stage of mineralisation. The superinduced content of sodium may then have originated in connection with the sodium replacement unit, representing the main influx of this alkali during the development of the pegmatite. An initial phase of this unit is supposed to consist of a saccharoid albit which in its main distribution occurs in the peripheral parts of the pegmatite. As this is just the location where varulite is found, one might assume that solutions in connection with the deposition of the saccharoid albite have contributed the content of sodium partaking in the metasomatic conversion of lithiophilite to varulite. The content of calcium partaking in the chemical composition of varulite is, however, still to be accounted for. The amount of CaO varies between 2.30 and 4.80 % in this mineral in strict proportion to the content of Na₂O (cf. analyses Table 15). An analysis of the saccharoid albite shows only 0.03 % CaO and cleavelandite in normal development 0.00 % CaO. The content of calcium in varulite can therefore hardly be ascribed to direct influence of solutions in connection with the sodium replacement unit in any other way than that they may have provoked reactions within the mineral assemblage they have replaced and thereby extracted a certain amount of calcium, subsequently influencing the metasomatic conversion of lithiophilite to varulite.

2. The caesium replacement unit

The large masses of pollucite in the Varuträsk pegmatite seem to call for a separate unit of replacement within the high temperature phase of the pneumatogenic stage. It seems hardly plausible that solutions of the same phase in some parts of the pegmatite have carried lithium as the main alkali component and close by have deposited great amounts of caesium in the form of the mineral pollucite. The localised distribution of the largest deposit along the core margin in the eastern wing likewise seems indicative of new replacement channels. The solutions must there have to a great extent replaced the mineral assemblages of the intermediate zones of the



Fig. 56. Large crystal of microcline perthite (M), successively replaced by spodumene (S), petalite (P), and pollucite (Po). Ultimately the whole mineral assemblage locally replaced by cleavelandite and quartz. Quarry P. Photo G. Gustavsson.

pegmatite stage. The quartz core, has however, not succumbed to any notable extent to replacements by the invading solutions of this unit.

Besides the accumulated masses of pollucite in the eastern wing (around the quarry P) the mineral also occurs, though in minor quantities, south of the large petalite area in the western wing (around F). The pollucite in this locality attracts attention through its pure white colour and vitreous lustre. In the mineral description specimens have already been denoted in which large crystals of bright red tourmaline are found enclosed in the clear white pollucite (see Plate I). As the two minerals must here have crystallised simultaneously, the red tourmaline, though in main pertaining to the subsequent sodium replacement unit, locally also appears in this unit, though only in connection with the occurrence of pollucite in the western wing.

In general pollucite occurs in the form of bulk replacement with few indications of sequence regarding the deposition. In a few places, however, evidences in this respect can be observed. On the clean-faced rock north of the shaft pollucite has been observed locally replacing petalite (cf. p. 120 and Pl. V, Fig. 2), indicating that the two minerals pertain to different phases of deposition. The sequence of mineralisation which can be observed in Fig. 56, leads to the same conclusions. A large crystal of microcline perthite (M) is there seen successively replaced by spodumene (S) and petalite (P) and ultimately by pollucite (Po). The whole mineral assemblage has subsequently been invaded by cleavelandite and quartz. It has been noted in another connection that petalite and pollucite have not been observed in direct contact with one another. So is also the case in this instance. In the photograph they are seen separated by the microcline crystal which they have successively replaced.

That pollucite belongs to a phase of mineralisation preceding that of the sodium replacement unit is clearly manifested by the circumstance that the large masses of



Fig. 57. Lepidolite pertaining to the polyolithionite series, partially replaced by cleavelandite (C). Near point F. $\times \frac{1}{2}$. Photo O. Mellis.

pollucite in the eastern wing are in all directions traversed by broader or narrower veins of albite. (See Fig. 34, p. 75.)

The only other mineral of any significance pertaining to this unit is a light grayish-white lepidolite, occurring in relatively coarse-grained masses around the same area to which the pollucite is restricted in the western wing. The fact that this is the only occurrence within the pegmatite of a lepidolite pertaining to the polyolithionite series may further testify its paragenetic association with the localised deposition of pollucite in this area. That this mica pertains to a phase of mineralisation, preceding that of the sodium replacement unit, is shown by the later invasion of cleavelandite in the form of numerous small fans (Fig. 57).

The observations now recorded coincide in testifying that the deposition of pollucite pertains to a younger phase of mineralisation than that of the lithium replacement unit, but to an older phase than that of the sodium replacement unit. This would confirm the supposition that the deposition of pollucite and connected minerals represent an independent replacement unit within the high temperature phase of the pneumatogenic stage indicating a break in the continuity of replacements during this phase.

c. The lower temperature phase

1. The sodium replacement unit

A further break in the sequential mineralisation of the pegmatite has now occurred. This is expressively indicated by two circumstances. In the first place the solutions

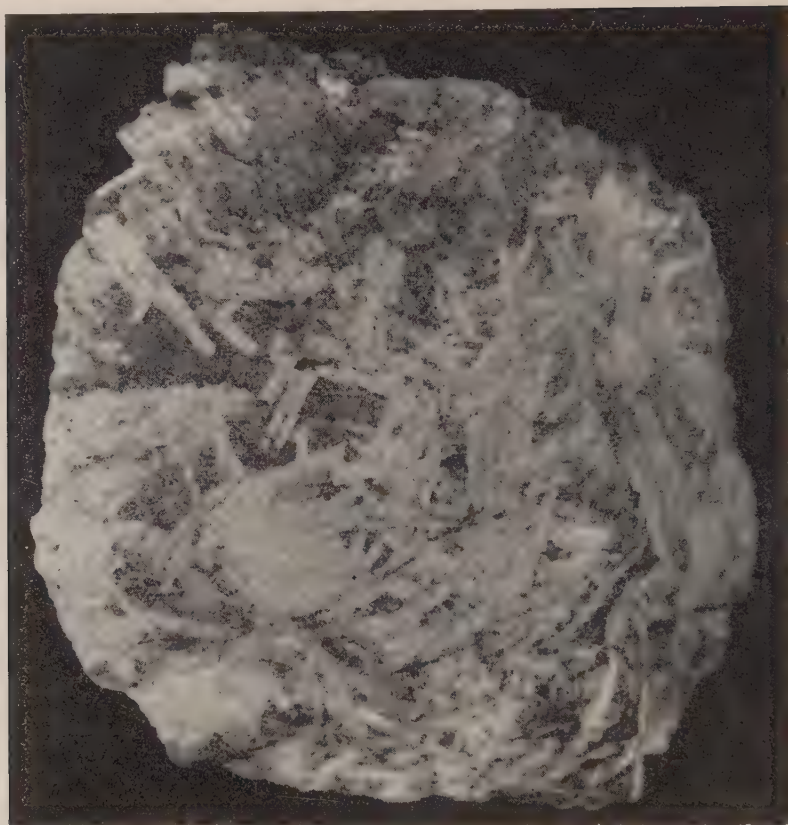


Fig. 58. Cleavelandite replacing dark vitreous quartz. Nat. size. Photo O. Mellis.

of this unit are universally found to traverse and replace all earlier consolidated mineral assemblages. Secondly this replacement unit has many new and characteristic features and includes minerals not before represented in the earlier zones or units. The difference in mineral composition is presumably due to the circumstance that the earlier solutions have to a large extent exhausted their content of the elements which characterised the minerals of the lithium and caesium replacement units. The residual solutions of the sodium unit were therefore lacking in these elements and enriched the others instead. With regard to alkalis, sodium now enters as the principal component. As a result of the influx of sodium, the dominant mineral of this unit is an almost pure albite, predominantly in the form of cleavelandite. In larger masses it is generally developed in spheroidal bursts or in large radiating sheaves. Fig. 39 gives a good illustration of such a cleavelandite burst, though only developed in a partial fraction. In Fig. 58 cleavelandite is seen replacing dark glassy quartz, where, as in other similar occurrences, it takes the form of more isolated laths. As already noted above in another connection (p. 79), the plagioclase also occurs in the form of a fine-grained saccharoid albite. This development seems to be principally restricted to more peripheral parts of the pegmatite, often reaching the wall zone of the pegmatitic stage as seen in Fig. 59.



Fig. 59. Saccaroid albite in contact with the quartz-muscovite-schorlite assemblage of the wall zone. Small needles of dark blue indicolite dispersed throughout the albite. Close to the hanging wall at G₁. Nat. size. Photo O. Mellis.

Next in importance of the minerals of this unit are different modifications of the mica minerals. The most abundant is a medium-grained gray lepidolite which, owing to a content of over 50 % muscovite radical, may be termed a lepidolitic muscovite (cf. analysis Table 26, No. 3). It often occurs as veins traversing any older mineral assemblage. In Fig. 60 it is seen intersecting petalite of a preceding unit. In other cases it takes the form of more accumulated bulk replacements and can

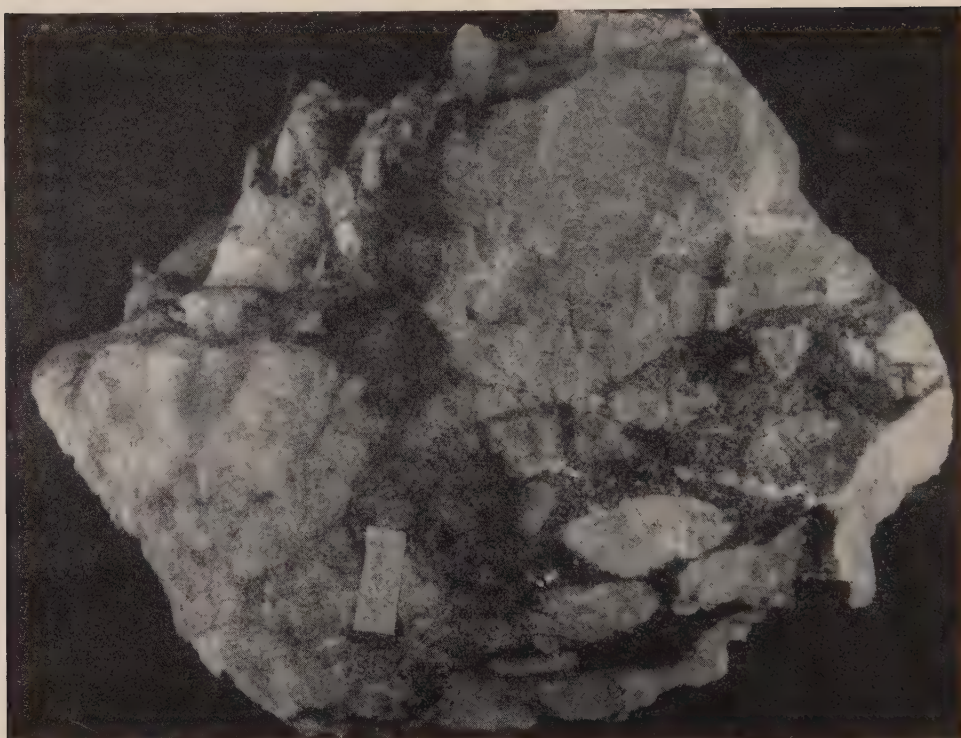


Fig. 60. Gray lepidolite pertaining to the sodium replacement unit, replacing petalite. $\times 8/5$.
Photo O. Mellis.

then be found locally replaced by cleavelandite indicating that this mica in parts has been deposited in advance of a concluding phase of cleavelandite deposition.

Another type of lepidolite of this unit is developed as large purple crystals, pictured in Fig. 32 (p. 71). This development is but scantily represented and principally restricted to the small prospecting pit T near the footwall of the pegmatite as well as in some large boulders near by. This lepidolite has a content of 5.95 % at the highest percentage found in any of the 12 analyses of the mica minerals Li_2O , Varuträsk, indicating a casual concentration of lithium in this place.

Other mica minerals, which belong to this unit, are a delicate rose-coloured to nearly colourless lepidolite, forming concentric bundles, and a rose-coloured fine-grained muscovite. The former is associated with cleavelandite and cassiterite around the excavation G₁. The analysis (Table 26, No. 4) showed an alkali content of 4.35 % Li_2O . This mica represents a true lepidolite, pertaining to the lepidolite series of Thelma Berggren (cf. p. 67, Fig. 30, No. 4). The other mica occurs in some abundance in and around the quarry P in the eastern wing. It contains only 0.69 % Li_2O and must be classified as muscovite with an insignificant content of lithium (analysis Table 26, No. 13).

Of other mica minerals pertaining to this unit may be observed a cryptocrystalline muscovite (oncosine), in general found traversing the large masses of pollucite around the quarry P and in the underground workings around the shaft but also occurring



Fig. 61. Cleavelandite replacing the fine-grained lepidolitic rock at the quarry H₂. $\times \frac{1}{2}$.
Photo O. Mellis.

in the form of independent replacements. These micas also only contain a small percentage of lithium (cf. Analyses 10 and 11 in Table 26) and must be named muscovites containing besides lithium, a certain amount of rubidium and caesium.

Indicolite and rubellite as well as the zonal tourmalines are characteristic minerals of this unit. Of the coloured tourmalines only rubellite has been found locally participating in a previous unit (see p. 104).

Manganapatite recurs in much the same aspect as found in the lithium replacement unit. It is one of the few minerals at Varuträsk which does not differ in habit though recurring in several different phases of mineral deposition.

Other minerals pertaining to this unit are a younger generation of lithiophilite now occurring in the form of small nodules in cleavelandite together with cassiterite, microlite and uraninite around G₁ (Fig. 64). To this assemblage also belong the rare minerals allemontite and stibiotantalite, only found around a small prospecting pit near G₁ in the western wing of the pegmatite. At least in general the tantalite minerals columbite and tantalite also pertain to the same mineral assemblage.

Some observations may now be given illustrating replacements during the consolidation of this unit. Both microcline, perthite and quartz of the pegmatitic stage as well



Fig. 62. Hanging wall and wall zone of the northern section of quarry P, partially replaced by cleavelandite and its associated mineral assemblage.

G, amphibolite; A, quartz-muscovite assemblage of the wall zone; C, cleavelandite; QC, quartz-cleavelandite intermixture; T, tourmaline (verdelite). Photo G. Gustavsson.

as petalite, pollucite and the lepidolitic micas of the preceding replacement units are almost everywhere found to be more or less intersected or replaced by cleavelandite and in more peripheral parts by saccharoid albite. Crossfractured green tourmaline is often healed by later intercalated albite.

Fig. 61 shows small radiating sheaves of cleavelandite branching off from narrow veins and replacing a massive gray lepidolite. In Fig. 52 (p. 99) the fine-grained lepidolitic rock of the lithium replacement unit was seen amply replaced by cleavelandite. In Fig. 62 large fans of cleavelandite are seen replacing the mineral assemblage of the wall zone.

In Fig. 63 fragments of a glassy beryl and of montebrasite, both in the modifications representative of the higher temperature phase, are seen encompassed by cleavelandite together with a brownish vitreous quartz and gray lepidolite. The beryl in the centre of the figure contains a small inclusion of montebrasite. The beryl and montebrasite fragments are surrounded by a rim of indicolite.

Fig. 64 shows nodules of lithiophilite pertaining to this unit together with casiterite and microlite, encompassed in cleavelandite.

A final illustration shows specific reactions involved during this phase. Nodules of montebrasite of the inner intermediate zone of the pegmatitic stage are there seen encompassed in cleavelandite though intercalated by a reaction rim of kaolinite and a micaceous mineral (Fig. 65). A coincidence of much the same nature has been described by Landes from Greenwood, Central Maine. Amblygonite nodules are there surrounded by crusts of kaolin. The illustration thereof in Landes' description (31, Pl. V) is in general aspect very similar to the occurrence at Varuträsk.

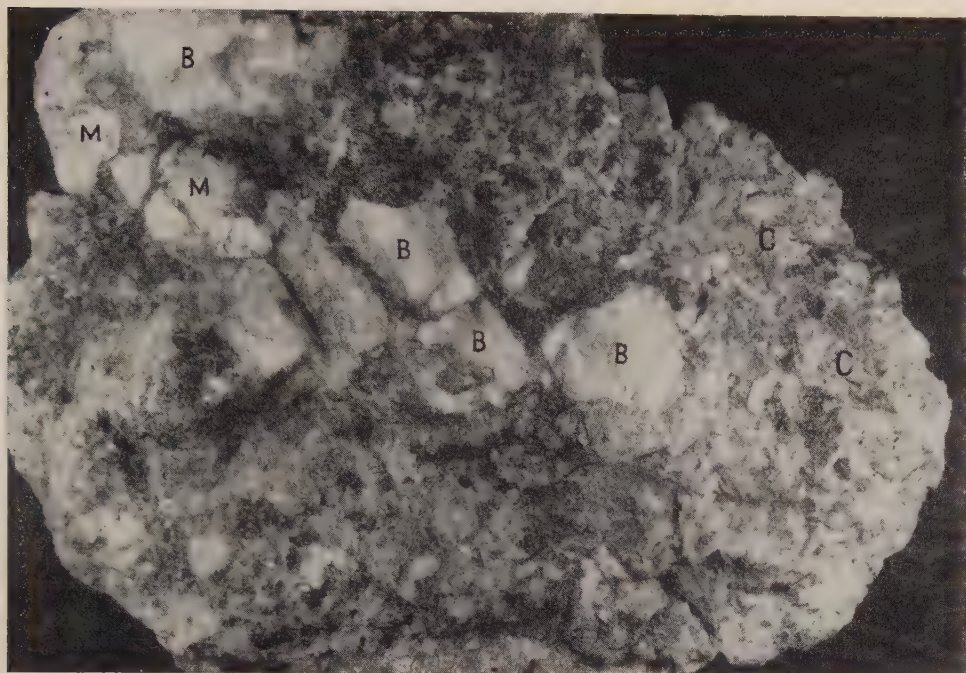


Fig. 63. Fragments of beryl (B) and montebrasite (M) encompassed in cleavelandite (C). The beryl in the centre contains a small inclusion of montebrasite. $\times 3/8$. Photo O. Mellis.

Summarizing the most characteristic features of the sodium replacement unit, the following points may be emphasized:

The dominant mineral of this unit is a chemically almost pure albite. It is most often developed in the form of spheroidal bursts or radiating sheaves of cleavelandite. Predominantly in peripheral parts of the pegmatite it is often substituted by fine-grained saccharoid albite which seems to have preceded the main deposition of cleavelandite. Both the feldspars invade all previously consolidated mineral assemblages of earlier stages and units.

The pronounced lithium-bearing silicates such as spodumene, petalite as well as pollucite are no longer represented in this unit. On the other hand lepidolitic micas of different aspects partake in the mineralisation during the whole period of mineral deposition pertaining to this unit. A gray lepidolite has partly preceded the main invasion of cleavelandite, but is also found in a concluding phase, subsequent to the consolidation of the main part of cleavelandite.

Of the coloured tourmalines, indicolite seems restricted to this unit as well as the zonal tourmalines. Rubellite, though occurring in the preceding caesium unit in a local assemblage, is commonly represented in this unit.

Lithiophilite recurs in a new development in the form of small nodules together with cassiterite, microlite and uraninite. The two latter minerals are definitely restricted to this unit as is also the case with the rare minerals allemontite and stibiotantalite.

The replacements during this unit have been very extensive, invading and replacing

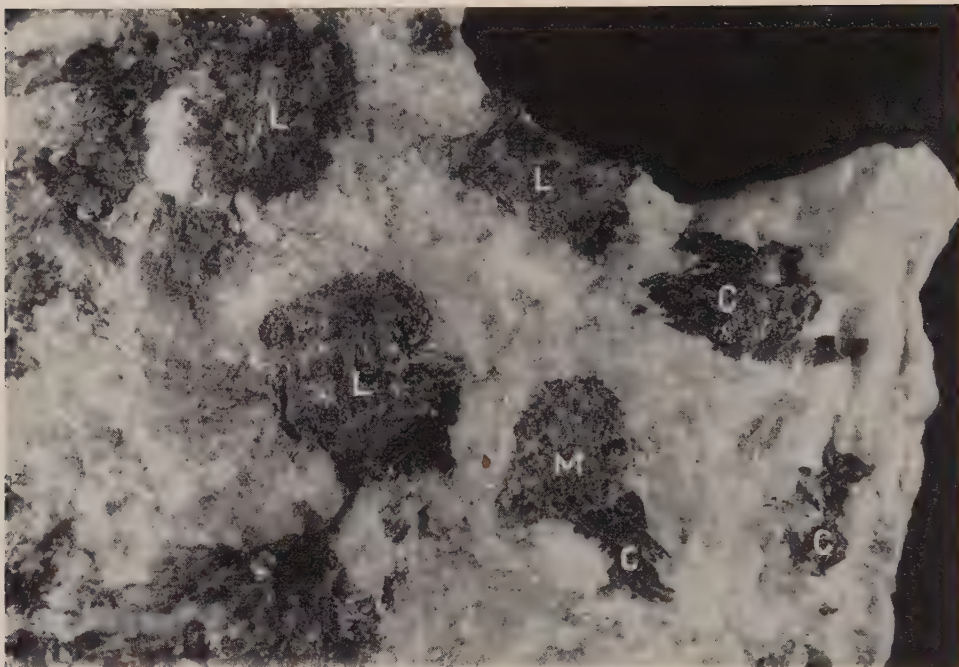


Fig. 64. Nodules of lithiophilite (L) together with cassiterite (C) and microlite (M) encompassed in cleavelandite. Excavation G₁. $\times \frac{3}{2}$. Photo O. Mellis.

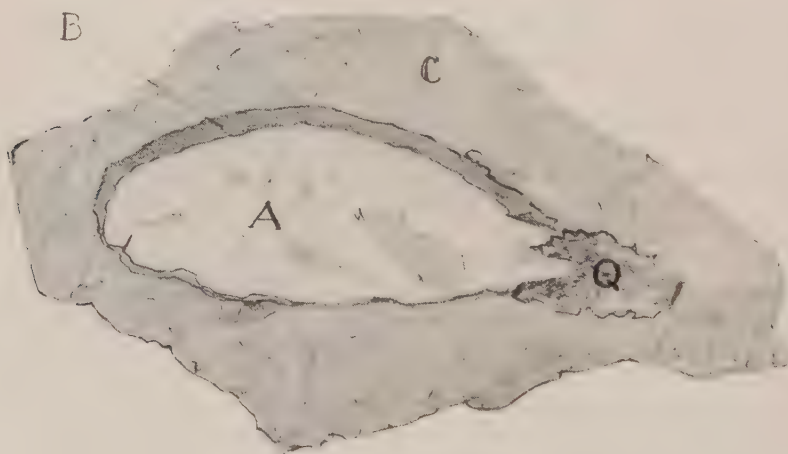


Fig. 65. Nodules of montebrasite (A) in cleavelandite (C) with reaction rim of clay minerals (B). Q, quartz. Nat. size. M. Rosenberg del.

large masses of all earlier mineral assemblages. This is especially the case in the western wing of the pegmatite (see Pl. V, Fig. 1). Most of the rare minerals pertaining to the sodium replacement unit are therefore only found in the western wing.

2. The fluorite unit

The reason for including a fluorite unit as a separate phase of mineralisation is that during five years of mining activity this mineral has only been found in two small vugs together with black vitreous quartz without any paragenetic connection with other minerals. As vugs are all but absent in the Varuträsk pegmatite, the formation thereof may imply a declining and concluding phase of mineral deposition of the pneumatogenic stage, in which final residual fluids have retained some fluorine and precipitated fluorite in the few vugs which occur. In the mineral descriptions in this paper it has been noted that such minerals as mangan-oxyapatite (mangan-voelckerite), mangan-hydroxylapatite and montebasite suggest that at the time of their deposition there was a deficiency of fluorine and chlorine in the mineral deposition. Mason concludes "that at no time during the deposition of the minerals of the pegmatite was there any excess of fluorine present" (45, p. 282).

The fluorite in the vugs occurs in the form of much etched colourless crystals.

4. The hydatogenic stage

The mineralisation of the hydatogenic stage is taken to represent all processes, which may be ascribed to the influence of ascending hydrothermal water percolating throughout the consolidated mineral assemblages of previous stages. The decomposition of selective minerals and therewith connective deposition of new minerals would represent a last phase of activity in connection with the paragenetic deposition of the pegmatite. Mineral decomposition due to superficial weathering is referred to a following heading.

During this stage the juvenile water has not introduced new material of any importance. The residual fluids of the preceding stage have apparently concluded the transfer of soluble matter.

The most pronounced feature of the minerals pertaining to this stage is their high content of hydroxyl radical, both in the form of absorbed and capillary water.

A characteristic mineral of this stage is cookeite, found replacing tourmaline as well as spodumene, where this mineral occurs in the same assemblage. An analysis (Table 28, p. 72) of cookeite in these connections shows a content of 12.40 % H_2O^+ .

The replacement of tourmaline is always restricted to rubellite. In most cases it affects the red kernel of the zoned tourmaline. One may then under the microscope follow every stage from a partial replacement with relicts of rubellite still preserved to complete pseudomorphs (cf. p. 64).

Another form of cookeite occurs as an independent mineral deposition, in its distribution coinciding with the latest generation of gray lepidolite of the preceding unit. The lower temperature prevailing during the hydatogenic stage would now favour the formation of cookeite rather than the less hydrous lepidolitic micas.

It is assumed that most other hydrated minerals in the Varuträsk pegmatite also belong to the mineralisation of this stage. This is probably the case with the rare phosphate mangan-hydroxylapatite, described by Mason (45, p. 380). He came to the conclusion that this mineral most probably replaces varulite. The hydroxylapatite contains 2.56 % H_2O^+ against about 1 % in varulite. On the other hand its oxidated

condition might indicate its pertinence to the stage of superficial alterations, were it not for the fact that no other indications of such nature are to be seen in the surrounding minerals than a thin coating of alluaudite on incidentally exposed parts of some varulite individuals. This form of oxidation is, however, equivalent to the alteration of triphylite into minerals of the sicklerite—heterosite group, which represents a different form of oxidation pertaining to the stage of supergene alterations.

Montmorillonite, when not intermixed with other clay minerals, is at Varuträsk only found as an alteration of petalite (cf. p. 73). The formation of this mineral is in general attributed to supergene decomposition. But when occurring in pegmatites it has however also been assigned to alterations of hydrothermal origin. Ross and Hendricks denote that montmorillonite, stable over a rather wide range of temperature, is formed by low-temperature hydrothermal processes as well as by weathering (58, p. 69). Concerning the formation of montmorillonite and kaolin they say: "Genetic processes taking place under hydrothermal or vapour conditions, are commonly characterised by the introduction of bases, including magnesium and iron. Where these bases are introduced, minerals of the montmorillonite group tend to form, where they are removed, kaolinite forms" (l.c. p. 79). Concerning montmorillonite from a pegmatite near Pala, San Diego, Cal., Schaller believes also that it was probably due to a late hydrothermal process (quoted by Ross and Hendricks p. 70). At Varuträsk all observations support the supposition that in this pegmatite montmorillonite in the main is of thermal origin, i.e. in the classification here applied, belonging to the hydatogenic stage. We must then, however, according to Ross and Hendricks, assume that the hydrothermal waters have assimilated a certain amount of magnesium and iron during their circulation before reaction with the petalite, as the latter mineral according to the analyses has 0.00 % MgO and only 0.20 % Fe_2O_3 against 4.54 % MgO and 1.54 % Fe_2O_3 in montmorillonite (cf. analyses Table 29). FeO is nil in both analyses.

Another incident tending to show that the formation of montmorillonite is not a product of supergene decomposition at Varuträsk is that it is in no way limited to such parts as could be assumed to have been affected by such processes.

What has been said above regarding the formation of montmorillonite is in many respects also applicable to the formation of kaolinite and other minerals of the kaolin group. In this case, however, no other interchange of material need be taken into consideration than an extraction of alkalies.

The younger generation of spodumene, belonging to the lithium replacement unit of the high temperature phase of the pneumatogenic stage is often found completely altered to kaolinite or to other clay minerals of more uncertain composition. Where the decomposition has reached completion, the final product can be scooped out by hand. Fig. 29 (p. 65) shows a broad vein of spodumene in microcline perthite. The upper part is quite fresh whereas parts of the lower half are fully decomposed. This and other similar occurrences are not indicative of supergene decomposition.

A transition may, however, have taken place between the mineralisation of the hydatogenic stage and that of supergene alterations. A decomposition initiated under hydrothermal conditions may continue in the form of supergene decomposition. Regarding the formation of kaolin and montmorillonite Noll says: "Was den Temperaturbereich der Kaolinbildung betrifft, so weisen Synthese und Paragenese in der Natur darauf hin, dass Kaolin ein mittel- bis niedrig thermales sowie epigenes Produkt ist. — — Bezüglich der thermodynamischen Parameter der Montmorillonit-

bildung gilt wiederum, in Übereinstimmung mit dem Experiment, das gleiche wie bei Kaolin" (49, p. 232, 234).

Eskola expresses the same line of thought when he says with regard to feldspars: "Aus dem Bereiche der hydrothermalen Metasomatose der Feldspäte leiten diese Vorgänge zu der hydrolytischen Spaltung der Silikatminerale bei der Verwitterung über" (11, p. 390).

It is probable that at Varuträsk the decomposition of spodumene to kaolin minerals has initiated under the hydrothermal activity of the hydatogenic stage but may have continued along similar trends during the succeeding stage of supergene alteration.

The alteration of pollucite to kaolinite is of a somewhat different character. It has only been observed in one locality in underground working proceeding from the shaft. Large masses of fresh pollucite were first disclosed. Twenty meters below the surface, along the contact between a large pollucite mass and adjacent mineral assemblages, mainly consisting of quartz, microcline perthite and petalite, the pollucite was, however, found to have succumbed to an intense alteration to a soft white clay substance.

As pollucite, when exposed to superficial weathering, as has been observed in several cases at Varuträsk, shows only insignificant indications thereof, the intense decomposition underground must in all probability be attributed to thermal activity even if decomposition also in this case may have continued during the stage of supergene alterations.

5. The stage of supergene decomposition

The essential feature regarding alterations during this stage is an oxidation, partly in connection with continued hydration, of selective minerals due to superficial weathering or to the action of percolating phreatic water.

A good example of such processes is the successive oxidation of triphylite and lithiophilite. In an intermediate stage the iron becomes trivalent, whereas manganese remains bivalent. Ultimately both the iron and manganese ions become trivalent in the fully oxidised end products heterosite and purpurite. Identically the same process of oxidation is the successive change of varulite to alluaudite-purpurite (cf. p. 50).

Arsenostibite (arsenian stibionite), an oxidation product of the alloy allemontite, must also be considered as an alteration pertaining to this stage. An obvious change in the relative proportions of Sb and As in allemontite and in arsenostibite may indicate that an initial substitution of the elements has taken place during the preceding hydatogenic stage. As As_2O_3 is more soluble than Sb_2O_3 in hot water, the percentage reduction of arsenic in arsenostibite may, however, be due to the juvenile thermal water of that stage.

Even if the initial formation of kaolin minerals as alteration products of spodumene and pollucite have been referred to the hydatogenic stage, it is possible that, as noted above, a continued formation of kaolin minerals during this stage may have taken place along mutual channels.

Besides the alteration of pollucite in the underground workings, noted above, a decomposition due to purely superficial weathering can be observed on exposed outcrops and in stray boulders. The mineral then shows a characteristic yellowish

coating. The veins traversing the pollucite then stand out in relief on account of their less susceptibility to weathering. Similar features have recently been described by Shaub and Schenck from Lithia pegmatite in Massachusetts (66, p. 661). The weathering was there restricted to a single boulder of pollucite. The authors assume that this apparently represents "the first instance where the effect of recent weathering on pollucite has been noticed". If this is the case, Varuträsk offers a second example. In both cases the pollucite under a thin weathered surface was perfectly fresh.

Fisher relates a singular occurrence of purely superficial weathering of pollucite from Tin Mine, South Dakota (25, p. 459). During mining the workers observed how fresh pieces of pollucite left on the ground after a few days became altered to a soft clay-like substance, easily scraped off by hand. (Personal communication.)

There hardly remain any further alteration products pertaining to this stage than a frequent incrustation of manganese oxides on minerals containing manganese in their composition or on adjacent minerals. It must, however, be taken into consideration that glacial erosion may have removed many products of weathering on exposed outcrops which otherwise might have increased the mineral assemblage of this stage.

V. Affinities with other pegmatites

Fennoscandian pegmatites

The Utö pegmatite in the archipelago of Stockholm bears comparison with Varuträsk in as far as it contains, besides the usual lithium minerals spodumene and lepidolite, also petalite in some abundance. The occurrence is of insignificant dimensions and all mining has long been suspended.

The Viitaniemi pegmatite (Eräjärvi, Finland) in many respects presents similarities with Varuträsk. It has a pronounced zonal development. In later replacement units a rich array of minerals occur, of which the phosphates have recently been described by Volborth (73 d). Many minerals are in common to both pegmatites and their sequence of deposition shows in many respects striking coincidences. It is of special interest to note that varulite here also has been identified as an alteration product of lithiophilite (l. c., p. 41).

Incidentally it may be observed that varulite has further been identified by Mason from the Skruppetorp pegmatite in Sweden (43, p. 178), and on account of the presence of its characteristic oxidation product alluaudite, it probably also occurs in the Sukula pegmatite in Finland (42, p. 372).

Several other pegmatites in southwestern Finland also offer similarities with Varuträsk in their mineral assemblage.

From the pegmatites in the Tamela district Mäkinen has notified the occurrence of, *inter alia*, triphylite and heterosite (73 a, p. 95).

From around Kimito Eskola records triphylite from a pegmatite at Skogsböle (73 b, p. 34) and Pehrman assumes that a black mineral there, analysed by Lokka, is arrojadite (73 c, p. 39). A sketch-map of Eskola (73 b, p. 39) in many respects reminds of the paragenesis of Varuträsk. The distribution of microcline perthite and accumulated quartz corresponds in detail to the zonal development

of the pegmatitic stage in the Varuträsk pegmatite, i.e. to its intermediate zone and core quartz. In that case the distribution of albite at Skogsböle might represent a core-margin replacement, in sequence pertaining to the pneumatogenic stage in the Varuträsk pegmatite (see Pl. IV).

From the Lemnäs pegmatite in the same vicinity Pehrman notes the occurrence of alluaudite which would signify the presence of its parent mineral varulite. Amongst other minerals in common with Varuträsk, montebrasite and cookeite may be mentioned as indicating further similarities of interest in the mineral assemblage of these two pegmatites.

Lithium pegmatites in other continents

Among other pegmatites which in respect of their paragenesis have recently been described more in detail, many from the North American continent present close affinities with the Varuträsk pegmatite. Some parallels may be briefly considered.

According to Fraser's description of the Newry pegmatite in Maine (15, p. 349) many similarities with Varuträsk are obvious both in general development as regarding details in the successive deposition of the mineral assemblages.

The characteristic minerals in common for both pegmatites are the silicates beryl, green, red and blue tourmaline, spodumene in two generations, lepidolite in different modifications, cookeite, pollucite, microcline, cleavelandite, the oxides cassiterite, columbite, microlite, uraninite and quartz, the phosphates manganapatite, montebrasite, triphylite and heterosite. The beryllium phosphates (beryllionite and herderite) as well as the phosphates eosphorite and francolite, all more or less common at Newry, are not found at Varuträsk. These minerals are, however, all developed in pockets which are all but absent at Varuträsk.

Regarding the occurrence of pollucite in the Newry pegmatite Fraser remarks that "it occurs closely associated with spodumene and in such a manner as to appear to be replacing it" (p. 359). In the Varuträsk pegmatite the deposition of pollucite has also been taken to have succeeded the lithium replacement unit and locally found replacing spodumene, pertaining to that unit. This would indicate a coincidence in the successive deposition of these minerals in the two pegmatites.

This seems also to be the case regarding the mica minerals. In the Newry pegmatite Fraser notes that in addition to large plates of lepidolite there occur two generations of coarse- to fine-grained lepidolite. The earlier is mauve in colour whereas the later is grayish (p. 358). This has also in the main been found to be the case at Varuträsk.

The noted incidences without doubt indicate many close affinities between the two pegmatites. Deviations are however also present. Among the lithium silicates petalite, abundant at Varuträsk, is absent at Newry. From a paragenetic point a significant dissimilarity also seems apparent. Fraser assumes that cleavelandite has preceded the main deposition of spodumene and pollucite (p. 352, 536) whereas at Varuträsk these minerals are everywhere found replaced by cleavelandite. There is no doubt that at Varuträsk the widespread replacements of cleavelandite have occurred after the consolidation of the mineral assemblages of the high temperature phase of the pneumatogenic stage whith both spodumene and pollucite as characteristic ingredients.

According to the description by Jahns (30, p. 1078) the Pidlite pegmatite in Mora County, New Mexico, has also many obvious similarities with Varuträsk. With the exception of calcic or median albite participating in the mineral assemblage of the

border and wall zones, the zonal development is identical. Analogies also exist regarding the paragenesis in the replacement units, though the mineral assemblages are in many respects of a different nature. The three generations of micas, recognised in the pidlite pegmatite, are to all appearances very similar to the same at Varuträsk, both with regard to their paragenesis and to their individual development.

Other North American pegmatites which offer similarities with Varuträsk are: the Branchville pegmatite (Shainin 63, p. 329), the pegmatite of Buckfield and Greenwood (Landes 31, p. 355), the pegmatites of Eight Mile Park, Col. (Heinrich 20, p. 420), and the pegmatites of south-east Manitoba (Stockwell 68, p. 27, 69, p. 37). According to the descriptions of the authors, many details may be found which coincide with equivalent conditions at Varuträsk in respect of both the general development and of the sequent deposition of the different mineral assemblages.

Simpson, in a series of papers, has described many minerals from pegmatites in Western Australia. A list thereof include columbite, tantalite stibiotantalite, lithiophilite purpurite, montebasite, Cs-beryl, coloured tourmalines, spodumene with alteration products (in part signified as cymatolite), lepidolite, a cryptocrystalline mica and petalite with montmorillonite as an alteration product thereof. One may say that this mineral assemblage in detail coincides with that of Varuträsk inasmuch as all the named minerals occur also there. Furthermore some significant and characteristic alteration products are in common. Some notes may therefore be given in this connection.

Stibiotantalite was first described from alluvial deposits around Greenbushes and later in small quantities from a greisenised pegmatite nearby. Simpson assumes that tantalite has preceded the formation of stibiotantalite, "the latter being formed by interaction between antimonial solutions and pre-existing tantalite in later stages of pegmatitic history" (67, IX, p. 14). At Varuträsk the stibiotantalite has been found to be replaced by microlite in connection with an extraction of antimony (cf. p. 30). From Simpson's description one is not convinced that the same may not be the case at Greenbushes.

From a pegmatite to Wodgina Simpson describes a compact cryptocrystalline muscovite occurring in large angular boulders (67, IX, p. 10). He says "the mineral is devoid of all visible trace of scaly structure, being very dense, tough and sub-vitreous and so compact that it carves well". This description corresponds in every detail to the cryptocrystalline muscovite from Varuträsk for which the name oncosin was applied (cf. p. 70). Independently Simpson has used the same name "as the nearest analogy in nomenclature".

In the Londonderry pegmatites petalite occurs in so great quantities that Simpson assumes that in this respect the occurrence is "undoubtedly one of the most remarkable deposits in the world" (67, XI, p. 121). In quantity this mineral at Varuträsk probably exceeds that of Londonderry. Each occurrence may however still represent one of the two greatest masses of petalite yet known.

VI. Summary and conclusions

The Varuträsk pegmatite has a typical zonal structure in its first stage of development. This stage is referred to as *the pegmatitic stage*. The sub-divisions into different zones (the border zone, the wall zone, the intermediate zones and the core) denote

a successive consolidation from the border inwards. A continuous fractional crystallisation has occasioned disparities in the mineral assemblage of the different zones. A first generation of spodumene and montebrazite occur in the inner intermediate zone.

After the final consolidation of the zones of the pegmatitic stage an intermission has taken place before the next stage of mineral deposition. Successive invasions have then occurred causing extensive replacements of the zonal mineral assemblages and depositing new minerals, varying in composition in accordance with the composition of the invading solutions and with the prevailing temperature during successive mineralisation.

This second stage has been named *the pneumatogenic stage*, indicating that the mineral deposition has taken place under conditions of pneumatolytic activity. The term pneumatogenic has seemed preferential to pneumatolytic for denoting a distinctive pegmatitic phase of mineral deposition than is comprehended in the wider conception attributed to pneumatolytic mineralisation in other geological connections.

This stage is assumed to include all the replacement units in which residual fluids have conveyed new material, subsequent to the mineralisation of the pegmatitic stage. The ruling temperature interval, during which these replacements have taken place, is assumed to lie between the inversion point of quartz and the critical temperature of the aqueous solutions, i.e. between approximately 600° and 400°. Adjustments in both directions must, however, be anticipated, depending on a number of internal and external conditions such as, *inter alia*, concentration of the various phases and the vapour pressure of the fluids (cf. Turner and Verhoogen 71, p. 331).

The replacement units of this stage have been sub-divided into a high-temperature phase, in which the main enrichment of lithium and caesium are found, and a lower temperature phase, in which sodium is the dominating alkali constituent.

The high temperature phase has been further divided into a lithium-replacement unit and a caesium replacement unit. The major proportion of the lithium-bearing minerals in the pegmatite have been deposited during the former unit, predominately represented by the minerals lepidolite, petalite and a second generation of spodumene. During the latter unit large masses of pollucite and quartz together with a small local occurrence of polyolithionite represent the only essential minerals.

The lower temperature phase has been sub-divided into a *sodium replacement unit* and a *fluorite unit*, the latter of insignificant import.

The sodium replacement unit is characterised by the abundant occurrence of albite, mostly developed in the form of cleavelandite. A second generation of lithiophilite now occurs associated with cassiterite, microlite and uraninite. The rare minerals allemontite and stibiotantalite pertain also to this unit as well as, at least in part, species of the columbite-tantalite group. Lepidolite and other mica minerals recur though in another development than in the preceding units.

A fluorite unit has been assumed to represent a last and concluding phase of mineralisation, pertaining to the pneumatogenic stage. It is only represented by isolated finds of fluorite and quartz in two small vugs, which otherwise are all but lacking in the Varuträsk pegmatite.

Regarding the sequence of successive mineral deposition during the pneumatolytic stage some significant observations from the bare-laid rock surface NE of the shaft have been put at my disposal by Mr. Wirstam. They are as follows:

Microcline perthite occurs as distinct inclusions in spodumene and montebrazite.

The contacts between petalite and the later generation of spodumene indicate a contemporaneous crystallisation of the two minerals.

The contact between petalite and pollucite is generally sharp. Occasionally, however, some slight replacement of petalite by pollucite may be observed.

Pollucite and the later generation of spodumene are always separated by veins of quartz.

Albite, lepidolite and vitreous quartz are the youngest minerals of this locality. The quartz penetrates all the above-mentioned minerals.

As may be seen, these observations correspond in detail with conclusions independently formed from other parts of the pegmatite, on which the sequence of minerals deposition in the preceding description has principally been founded.

The third stage named the *hydatogenic stage* is taken to include all alterations of the mineral assemblages of preceding stages due to the activity of ascending juvenile water. The term *hydatogenic* is in accordance with the term *pneumatogenic*, as applied for the preceding stage, taken to signify the phase of mineralisation which has originated under conditions of hydrothermal activity. In conformity with Fersman, Schneiderhöhn and others, the conception hydrothermal is restricted to the mineral deposition which has taken place below the critical temperature of the aqueous solutions instead of extending it to include phases of mineralisation above this temperature, signified by others as high temperature and intermediate temperature hydrothermal phases, directly succeeding the epimagmatic stage of consolidation (cf. p. 85).

No essential addition of new constituents is anticipated to have occurred during this stage. The approximate temperature interval is assumed to lie between 400° and 100°. The characteristic feature of this stage, as far as the formation of new minerals is concerned, is that in general they represent hydrated forms of earlier formed minerals. Cookeite, representing a hydrated equivalent of the mica group, is a characteristic representative of this stage. Montmorillonite as an alteration of petalite and, at least in part, certain minerals of the kaolin group are also assumed to pertain to this stage.

The final *stage of supergene alteration* is in Varuträsk of minor importance. Probably most of what may have pertained to this stage has been effaced by glacial erosion. What remains is mainly restricted to oxidation products of the lithium-bearing phosphates, where these have been exposed to superficial oxidation. A coating of manganese oxide on or near manganese-bearing minerals naturally also pertains to this unit. A continued formation of minerals of the kaolin group may, as noted above, also be referred to this unit.

A striking feature of the Varuträsk pegmatite is the large accumulation of rubidium and caesium. The former is principally concentrated in the microcline perthite which can attain up to 3 % Rb_2O . Taking only an average of 1 % Rb_2O (as the content is obviously lower in the feldspar of the eastern wing of the pegmatite) and estimating the total amount of microcline perthite already excavated at around 3000 tons and quantities still unmined to at least some 1000 tons, this would represent over 30 tons metallic rubidium concentrated in the pegmatite. An estimate of the content of pollucite is at present not available but it is probable that Varuträsk contains the highest amount of this mineral in any known pegmatite, and as caesium also enters many of the mica minerals, the accumulated content of caesium is probably greater than in any other known pegmatite.

Some minerals worth special attention may be noted in this summary. The occurrence of stibarsen and other subspecies of the allemontite group are of interest

as these alloys have not previously been recorded in pegmatites. Av Varuträsk they have only been found in one small prospecting pit. Stibiotantalite has also only been found in one locality close by the find of allemontite. The proximity of the only occurrences of these rare minerals must indicate that, in this restricted area, a fortuitous concentration of antimony and arsenic has taken place in the residual solutions of the sodium replacement unit, resulting in the local deposition of these minerals.

Finally the occurrence of the new species varulite may be observed, representing an alteration product of lithiophilite, as well as of its oxidation products alluaudite and purpurite in accordance with the similar oxidation of triphylite and lithiophilite to minerals of the sicklerite group and to heterosite-purpurite.

In Plate II the sequence of the characteristic minerals of the Varuträsk pegmatite is outlined, as far as can be conjectured from observations in the field. It must, however, be kept in mind that the distribution of certain minerals may have a somewhat wider range than given in the table on account of secondary reactions between zones and replacement units, causing a transmission of minerals beyond their primary fields of distribution.

In the table the temperatures which are presumed to have prevailed during certain phases of mineralisation have only been indicated as temperature intervals on account of uncontrollable circumstances which may shift the temperature scale.

The primary zonal development of the Varuträsk pegmatite is assumed to have taken place in a closed system. The consecutive mineralisation of the different zones conform with what can be anticipated as the result of a fractional crystallisation without transmitted material from an outer source. The interchange of material within the zones of the pegmatitic stage can in every case be attributed to internal reactions within the same.

The pronounced variations in chemical composition within the mineral assemblages of the pneumatogenic stage can on the other hand not be assumed to have originated under conditions prevailing during an extended development within a closed system. The most plausible supposition must instead be that superimposed material has been transmitted from solutions, arising from genetically connected deepseated outliers of the pegmatite body. A successive and varied mineral deposition from such fluids would then intermittently be dispersed throughout the pegmatite and give rise to bulk replacements or fracture fillings.

The difference in chemical composition in the mineral assemblages of successive replacement units is essentially due to an interchange of the alkali components. In the pegmatitic stage these were predominantly, potassium and rubidium. Lithium, caesium and sodium sequentially prevail in the following phases of mineralisation.

A problem in connection with replacement is to account for the redeposition of removed material. In a paper on "Phases of mineralisation in Namaqualand pegmatites" Gevers has discussed this question in some details. He assumes that "many of the alterations taking place are not always replacements in toto but merely involve the interchange of certain radicals and the removal of only comparatively small portions of the replaced material" (16, p. 360).

With regard to conditions at Varuträsk this conception may be in part applicable in so far as most of the replacements, besides an interchange of alkalis, mainly involve an interchange of silicate radicals. Where, however, huge masses of the pegmatitic mineral assemblages have been replaced, as is the case in different parts

of the Varuträsk pegmatite, a certain amount of material must at all events have been removed (see Pl. V, Fig. 1). Beside alkalis this would include such silicates radicals which had not entered the newformed minerals. The same fluids which participated in the processes of replacement may then have retained in solution a certain amount of resolved material. The redeposition thereof has presumably taken place within the surrounding rock formations, through which residual fluids can have dissipated. There are no indications at Varuträsk that re-dissolved material has been retained within the pegmatite body.

In the mineral descriptions a preliminary determination of the age of the Varuträsk pegmatite was given, based on isotopic determination of the lead isotopes in uraninite by W. Wahl (p. 37). The age was calculated to be approximately 1.70×10^9 years. A determination with the $\text{Rb}^{87}\text{Sr}^{87}$ method by S. Eklund was given on the same page based on a new calculated half-life value of Rb^{87} , from which Eklund derived an age of 1.74×10^9 with a relative error of 12 % (10, p. 78).

These determinations tallied with each other as well as with preliminary determinations by Parvel and Wickman for the Ytterby pegmatite and related pegmatites at Digelskär in the archipelago of Stockholm and Kårarvet by Falun, with an age estimated to lie between 1.70 and 1.81×10^9 years (51, p. 354). These pegmatites are assumed to derive from granites geologically contemporaneous with the granite connected with the Varuträsk pegmatite.

On the other hand age determinations by Ahrens (2, p. 193) on lepidolite from Varuträsk gave essentially higher values. From an average of four determinations with the Rb/Sr method Ahrens calculated an age of 2.15×10^9 years, considerably deviating from the calculated ages given above. Recently, however, K. Fritze has published a paper with the title "Vergleich der Kalium-Argon-Metode zur geologischen Altersbestimmung mit der Rubidium-Strontium-Metode an identischem Material" (Dissertation, Mainz 1955). With the $\text{K}^{40}/\text{Ar}^{40}$ method a lepidolite from Varuträsk gave an age of 2.06×10^9 years and of the same lepidolite with the $\text{Rb}^{87}\text{Sr}^{87}$ method likewise 2.06×10^9 years. This seemed a singularly good correspondence. However, a determination on microcline from Varuträsk with the K/Ar method by Fritze executed in a similar way, gave an essentially lower age of 1.62×10^9 . He assumes that this may be due to argon loss through grinding the more brittle material, as anticipated by Waserberg and Hayden (*Geochimica and cosmochemica Acta* 7, 1955, p. 57), which would give a lower ratio $\text{Ar}^{40}/\text{K}^{40}$ and hence a lower age. Fritze however concludes that the disagreement in age still awaits a satisfactory explanation.

Recently it has been suggested in the United States that the assumed branching ratio of potassium and the assumed half-life value of rubidium may still be inaccurate (personal communication). In such a case it would be premature to give a definite age of the Varuträsk pegmatite. All that one can say with any degree of probability is that the age will be found to lie between 1.8 and 2.0×10^9 years, i.e. close to or between the ages calculated by Parvel-Wickman and Fritze.

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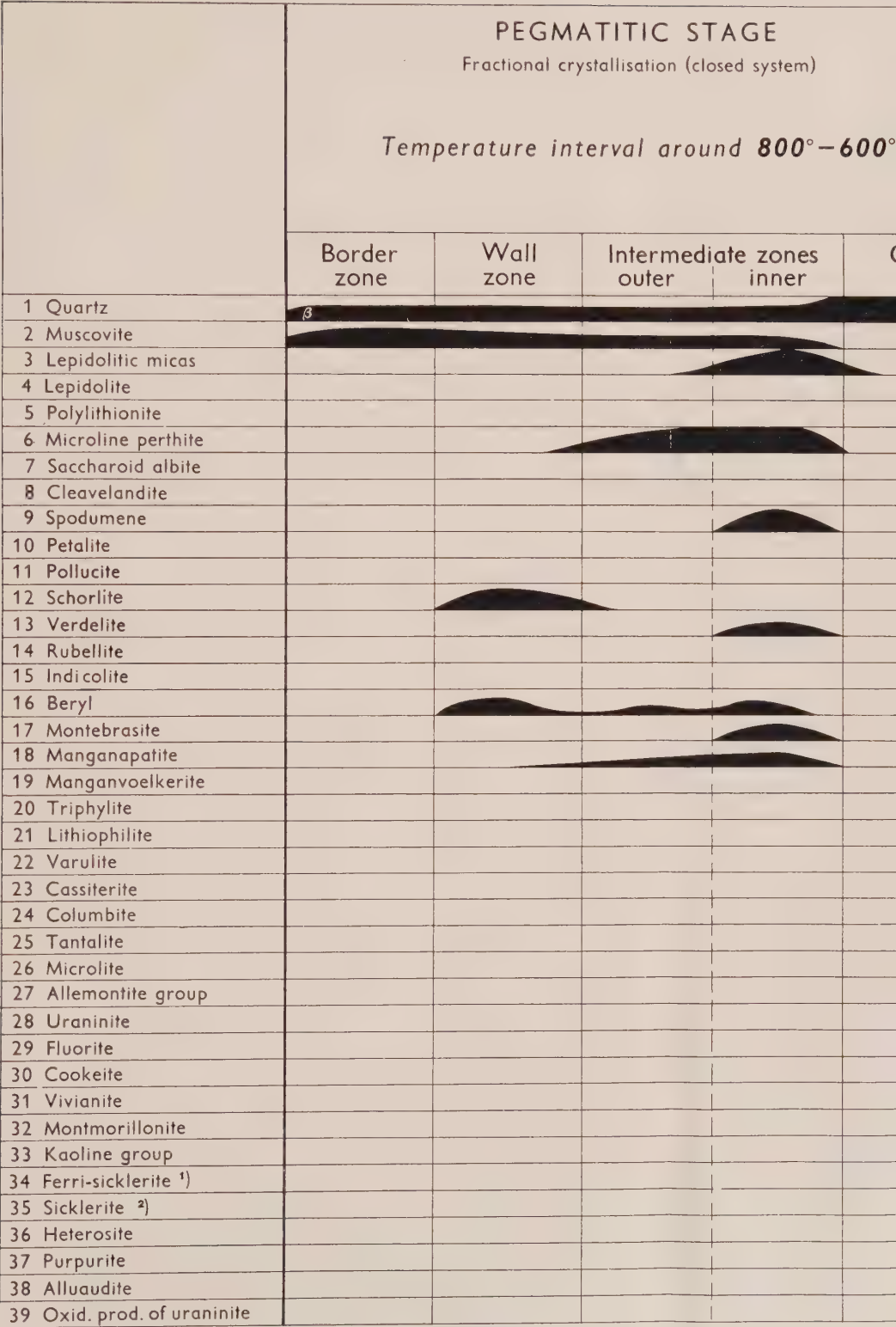
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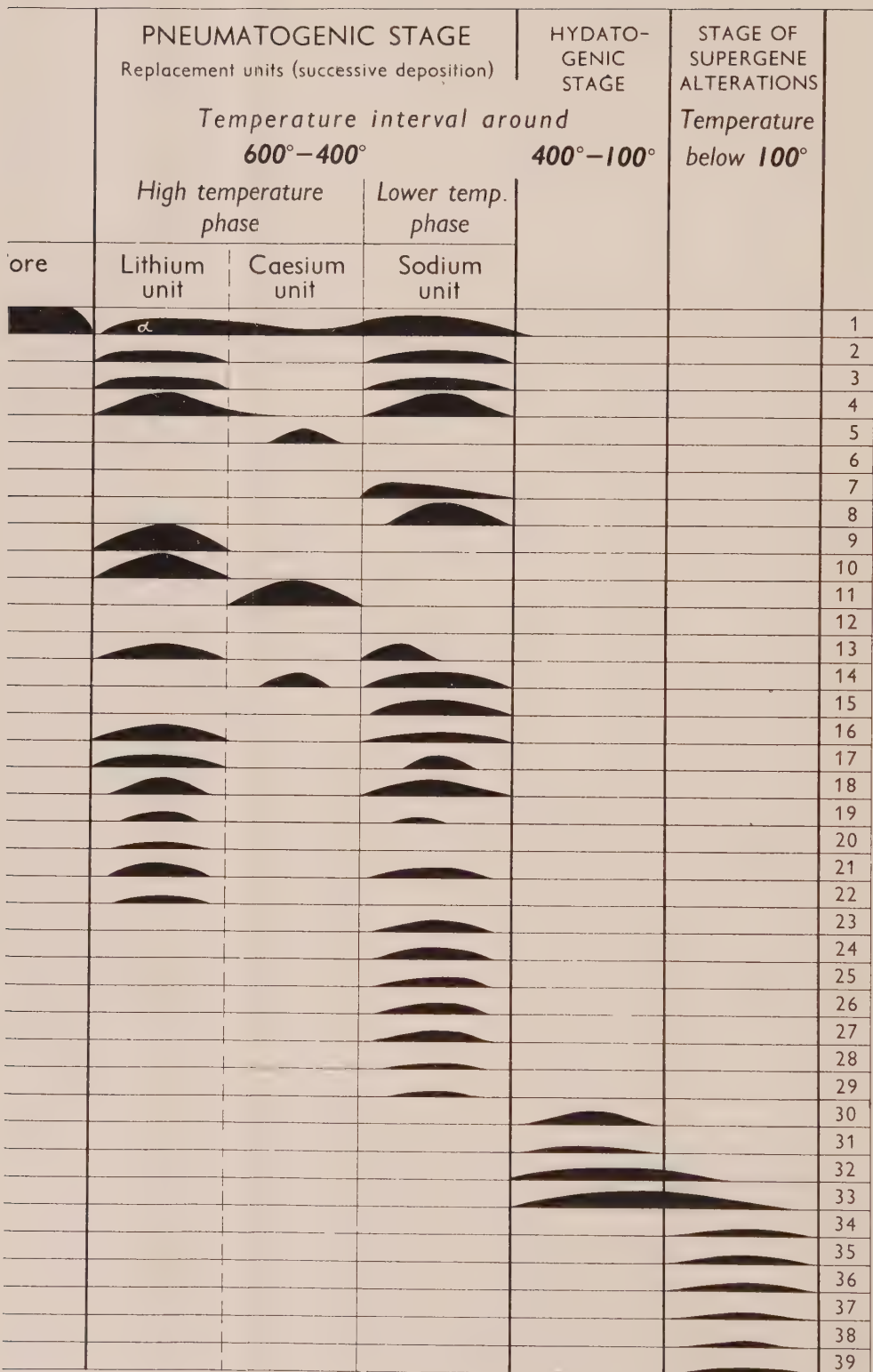
Pollucite intercalated with crystals of rubellite. Excavation around F. Natural size.

THE VARUTRÄS
Paragenesis of



¹ Ferri-sicklerite (ferrian sicklerite). ² Sicklerite (manganoan sicklerite).

PEGMATITE the minerals



Explanatory notes to the map (Plate III) and Plates V-VI

The map (Pl. III) is a surface plan of the exposed parts of the pegmatite. The north-eastern part, outwards from the dashed demarcation line on the map, is devoid of exposures and prospector drilling, except a small excavation around T. The contours limiting the phases of mineralisation within this part are therefore speculative.

The letters B to T on the map indicate the position of mining operations. Many of them are referred to in the text and in the illustrations as indicating the location for certain minerals or mineral assemblages.

All the drillholes made by the Boliden Company during prospecting work are plotted. Only the numbers 10, 22 and 39, signifying localities referred to in the text, have however been inserted. The shaft, marked on the map, has been sunk to 18.5 m level.

The outer and the inner intermediate zones of the pegmatitic stage have not been separated on the map on account of their frequently indistinct boundaries.

The replacement units of the pneumatolytic stage only include the two high temperature phases (the lithium and the caesium replacement units). The subsequent lower temperature phase (the sodium replacement unit) is so irregularly dispersed throughout the pegmatite—in general it is only present in minor quantities—that it has not been possible to carry through a satisfactory demarcation in the scale of the map. In Pl. V, Fig. 1, however, an unusually large bulk replacement pertaining to this unit can be seen.

Some significant instances referring to the location of certain minerals or mineral assemblages on the map may briefly be recapitulated. Fig. 2, Plate V shows the intricate replacements of minerals pertaining to the pneumatogenic stage at 35 m level around drill hole 39.

The largest masses of petalite are found at and south of D. The largest masses of pollucite are located around quarry P and at lower levels in the underground workings around the shaft (see Pl. VI).

The large beryls, pertaining to the wall zone of the pegmatitic stage, chiefly occur around H₁.

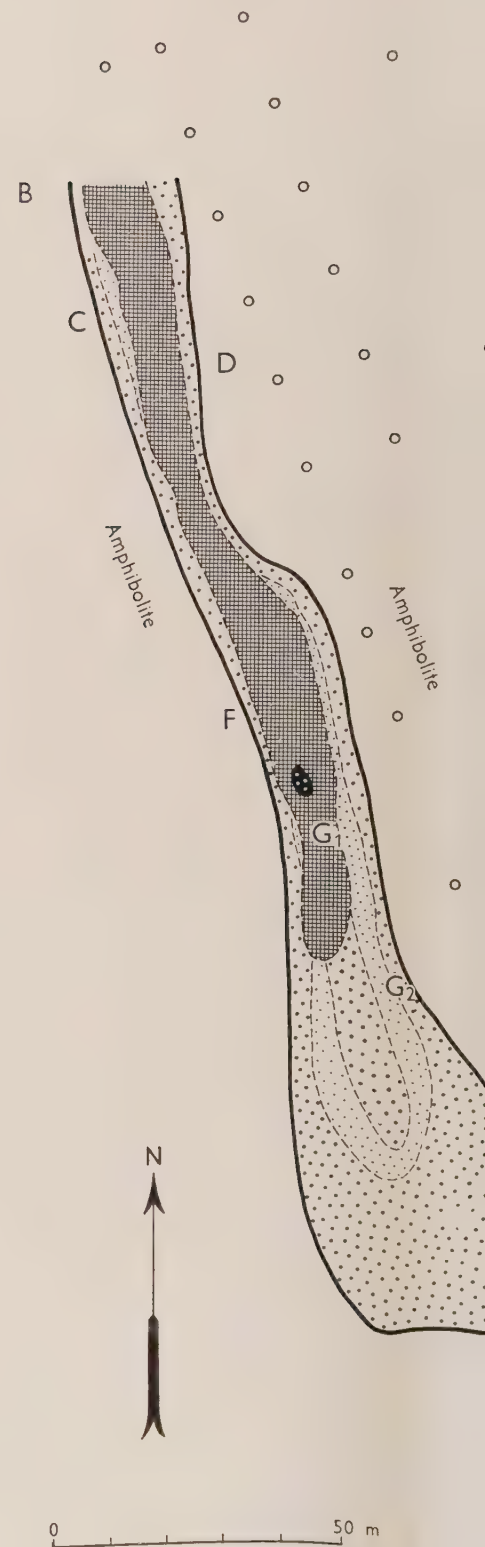
The large mass of fine-grained lepidolitic rock occurs around H₂.

The large crystals and anhedral masses of montebrasite, pertaining to the inner intermediate zone of the pegmatitic stage, occur chiefly around the quarry P and in the underground workings from the shaft.

The large toothshaped crystals of lepidolite as well as the finest zonal tourmalines are found at T and in large boulders nearby.

The only find of allemontite and stibiotantalite is located to two small excavations around G₂. The insignificant quantities of uraninite occur together with cassiterite and the younger generation of lithiophilite around G₁.

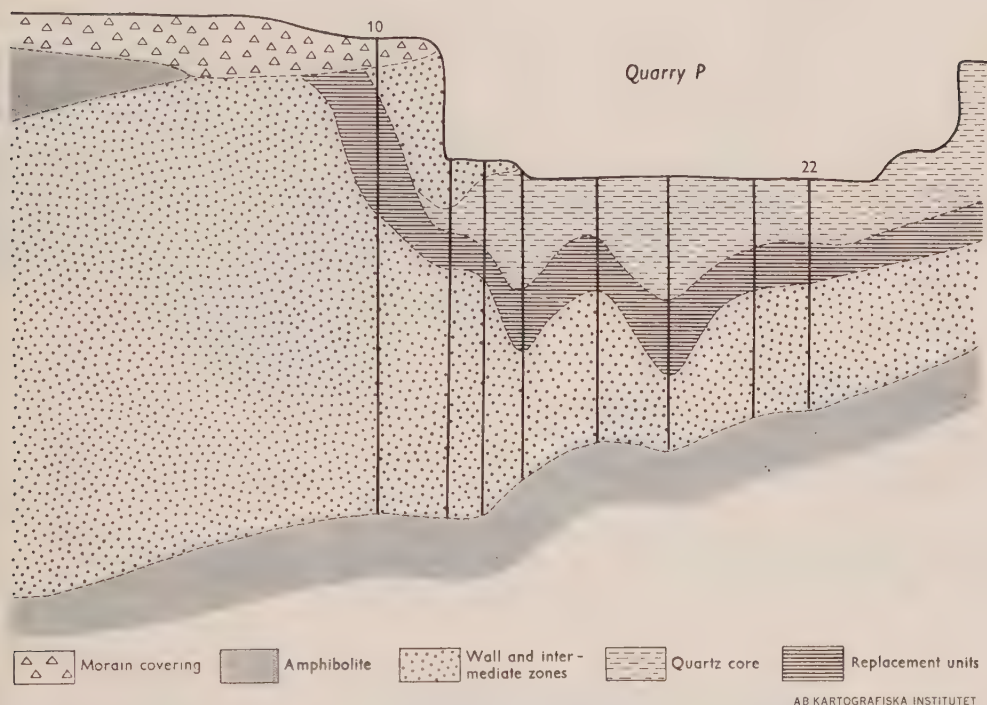
The clean-faced rock surface, often referred to in the text, is situated 30 m NE of the shaft (reproduced in Fig. 46).



THE VARUTRÄSK PEGMATITE

Surface plan showing zonal distribution and the two high-temperature replacement phases of the pneumatogenic stage (the lithium and caesium replacement units)





Vertical sections between the drill holes 10 and 22 on the map. Note the restricted core-margin situation of the replacement units in this area which mainly consists of lepidolite. In one of the drill holes an insignificant amount of pollucite was observed in contact with the core quartz and separated from the underlying lepidolite by about an equal amount of montebrasite.

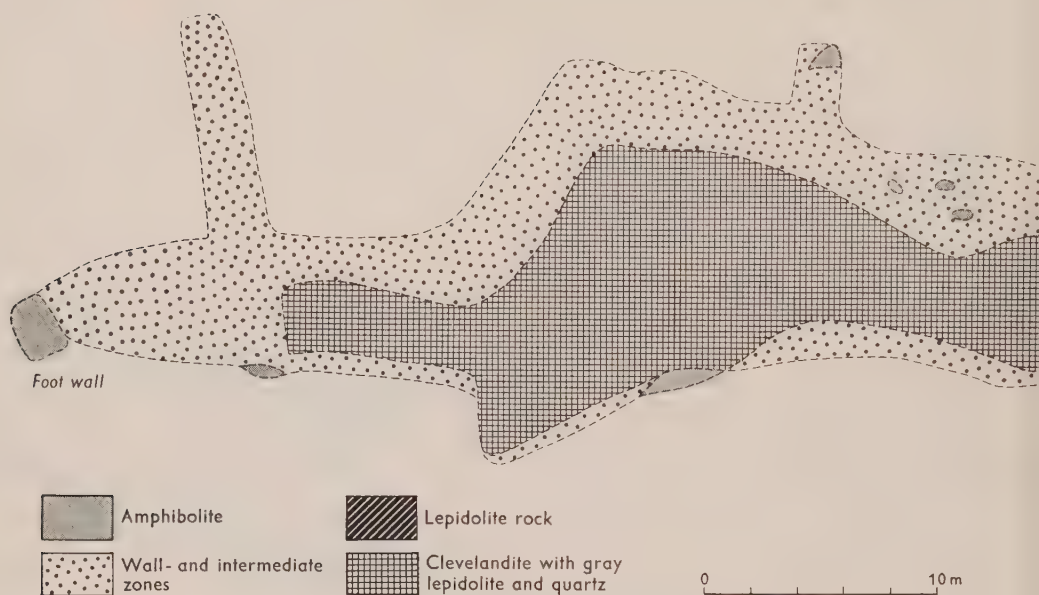


Fig. 1. Sketch showing extension of the massive lepidolitic rock around quarry H_2 , as well as a large replacement of cleavelandite and associated gray lepidolite throughout the mineral assemblages of the wall- and intermediate zone. Numerous xenoliths of the surrounding amphibolitic rocks are seen in the wall zone.

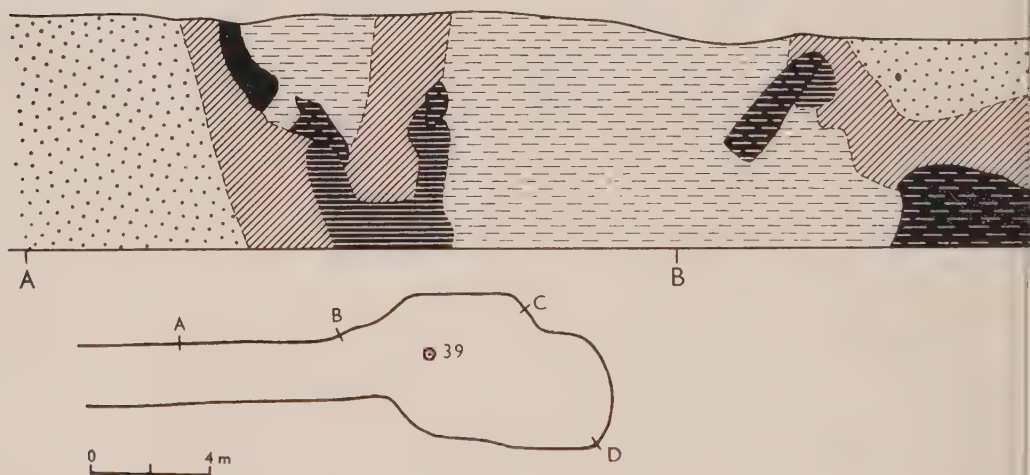
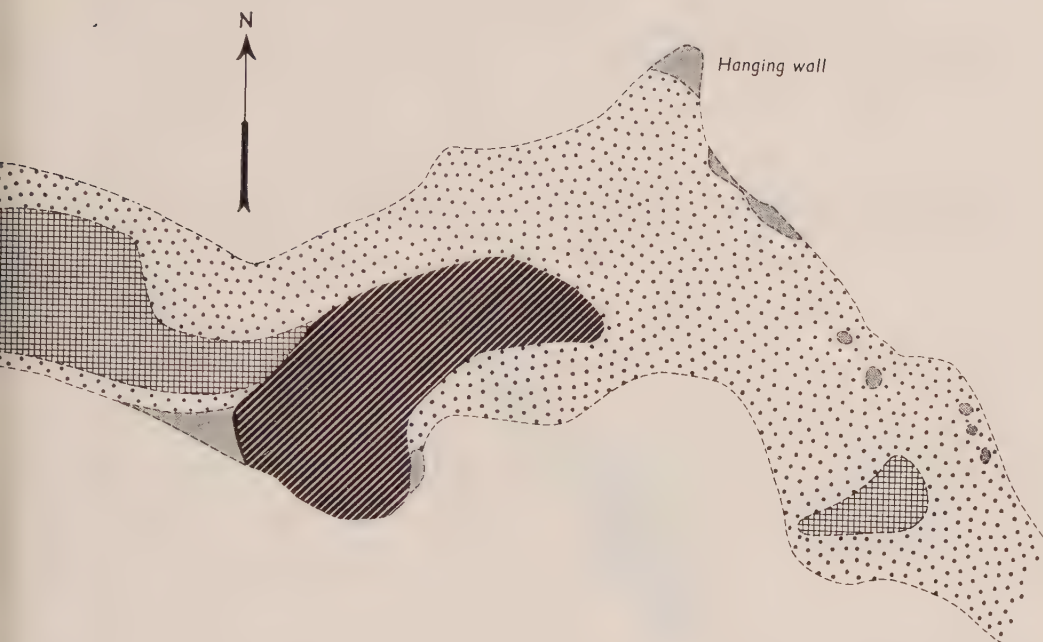
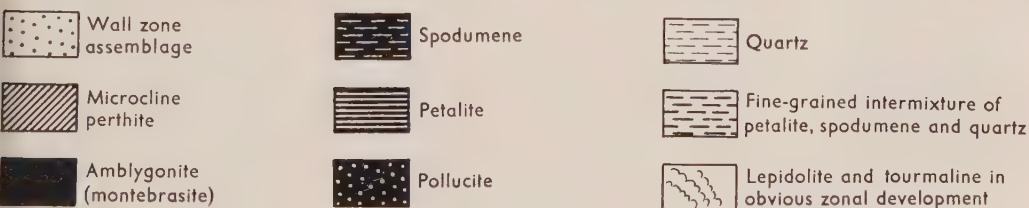
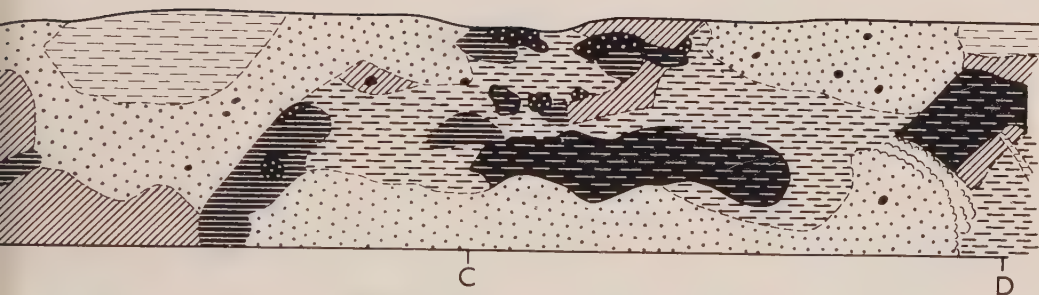


Fig. 2. Sketch by E. Grip showing distribution and paragenesis of the mineral assemblages of the pegmatitic and pneumatogenic stages at 35 m level around the drill hole 39, indicated on the map. To the left spodumene of the second generation and petalite are seen replacing microcline perthite. To the right pollucite is seen replacing petalite.

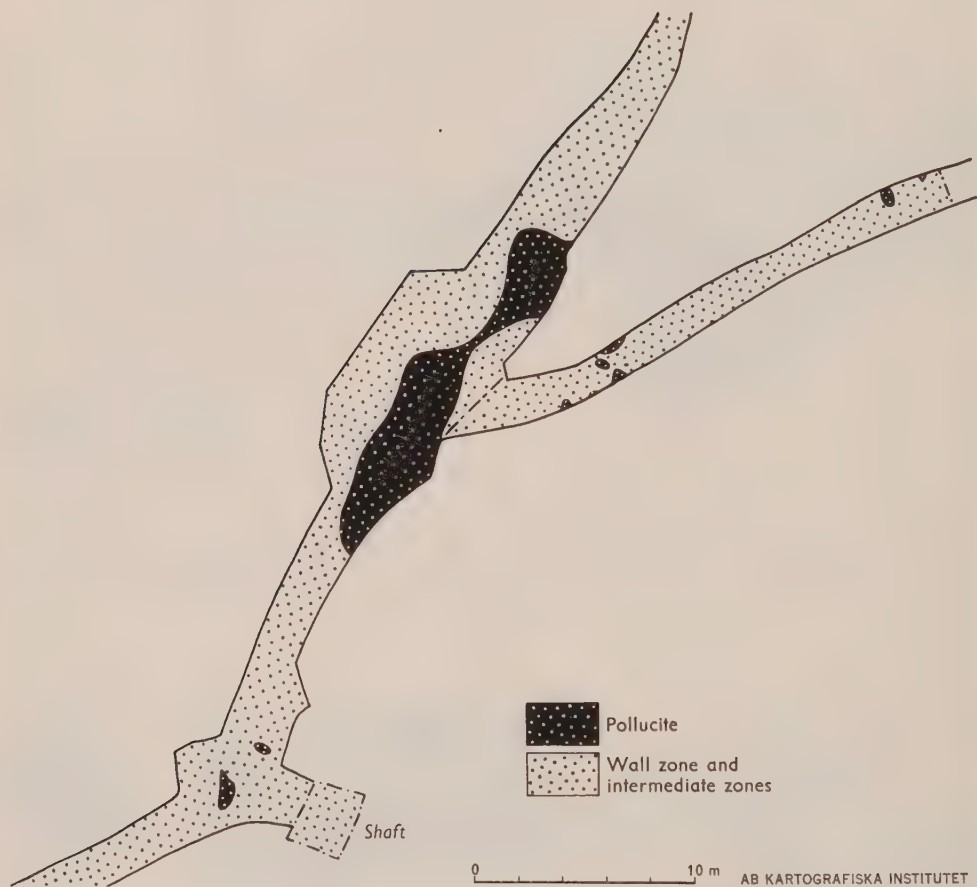


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Sketch showing extension of pollucite pods at 37 m level in the underground workings north of the shaft.

An iron-rich scorzalite from Hållsjöberget (Horrsjöberget), Sweden

By ÅKE HENRIQUES

Abstract

Scorzalite, $(\text{Fe, Mg})\text{Al}_2(\text{PO}_4)_2\text{H}_2\text{O}$ has been found in Sweden in the lazulite-bearing kyanite-quartzite at Hållsjöberget (Horrsjöberget) in Värmland. The nature of the occurrence, mineral association, chemical analyses, X-ray data and the specific gravity and optical properties of the mineral are described. Further, an account of a new chemical and optical determination of lazulite $(\text{Mg, Fe})_2\text{Al}_2(\text{PO}_4)_2\text{H}_2\text{O}$ from the same deposit is presented.

Introduction

Lazulite and scorzalite form an isomorphous series $(\text{Mg, Fe})\text{Al}_2(\text{PO}_4)_2\text{H}_2\text{O}$. While the mineral name lazulite has been in use for over a hundred years, the name scorzalite was proposed as late as 1947 by W. T. Pecora and J. J. Fahey as a term within the isomorphous series to be applied when the molecular ratio $\text{Fe}^{2+}:\text{Mg}$ exceeds 1:1. Scorzalite is previously known from only three localities, namely, Corrego Frio in Minas Gerais, White Mountain in California and the Victory mine in South Dakota. The scorzalite from Hållsjöberget has a $\text{Fe}^{2+}:\text{Mg}$ molecular ratio of 87:13, while the scorzalite from the Victory mine, the most iron-rich hitherto known, has a corresponding molecular ratio of 76:24.

The kyanite-quartzite at Hållsjöberget

The kyanite-quartzite at Hållsjöberget consists of a rather small quartzite occurrence in the Pre-Cambrian basement, and is interbedded in gneisses and fractured by hyperites. The quartzite is banded as a rule with alternating bands rich in quartz and kyanite respectively. The thickness of the bands varies considerably, but is usually about one or two centimetres. When the quartzite is not banded the content of kyanite seems to be much greater. Scorzalite, as also lazulite, occurs mainly in the kyanite-rich parts. The two minerals are generally congregated separately in fairly small masses together with, in particular, quartz, rutile and kyanite. Other minerals also occur here, and sometimes pyrite, haematite, ilmenite, apatite, tourmaline, micaceous minerals etc. may be observed. Small grains and granular aggregates (0.1—5 mm) of lazulite and scorzalite are also spread throughout the quartzite. No particular stratigraphical connection between the distribution of lazulite and scorzalite has been observed, although lazulite seems to be much commoner than scorzalite.

Table 1.

(Analyst A. Aaremäe)

	1.	Mol. prop.		2.	Mol. prop.	
SiO ₂	0.06	0.0010		0.20	0.0031	
TiO ₂	0.41	0.0051		0.31	0.0039	
Al ₂ O ₃	28.27	0.2773	0.2960 1.02	31.19	0.3060	0.3063 0.93
Fe ₂ O ₃	2.98	0.0187		0.04	0.0003	
FeO	17.70	0.2463		8.40	0.1169	
MnO	0.49	0.0069	0.2894 1.00	0.12	0.0017	0.3309 1.00
MgO	1.46	0.0362		8.56	0.2123	
CaO	0.62	0.0111		0.04	0.0007	
P ₂ O ₅	42.20	0.2973	0.2936* 1.01	44.74	0.3152	0.3152 0.95
H ₂ O ⁻	0.07	0.0039		0.10	0.0055	
H ₂ O ⁺	5.66	0.3441	0.3141 1.09	6.16	0.3418	0.3418 1.03
	99.92 %			99.86 %		

* 0.0037 units combined in apatite.

1. Scorzalite from Hållsjöberget.

2. Lazulite from Hållsjöberget.

Calculated mineral composition:

1. Scorzalite	98.27 %	2. Lazulite	99.21 %
Apatite	1.17	Quartz	0.20
Quartz	0.06	Rutile	0.31
Rutile	0.41		

Investigation and treatment of the material

The material for chemical analysis and other determinations was selected from 123 g of a scorzalite-rich sample. After the specimen had been crushed and sieved (+150 mesh, Tyler standard) separation by means of heavy liquids was carried out, after which the material was hand-picked under the microscope. The chemical analysis (Table 1) shows that in spite of the above precautions small quantities of impurities accompanied the analyzed sample. These impurities, mainly quartz, rutile and apatite, could, however, be determined qualitatively under the microscope and were later calculated quantitatively from the result of the analysis. The recalculated analysis seems, therefore, to give a reliable picture of the chemical composition of the scorzalite here described.

The refractive indices were determined by the immersion method. Other optical determinations were performed with Universal Stage. The optical angle was measured in mineral grains in which the emergents of both optical axes could be observed, i.e., within the optical axial plane (010) and in a section approximately at right angles to the α -direction. Sodium-light was used in all the determinations. The specific weight was calculated by the suspension method with the assistance of homogeneous grains of scorzalite (+50 mesh), temp. 20°C.

The determinations in the case of lazulite were carried out in the same manner. The material was obtained from a lazulite-rich sample weighing 98 g (obt. fr. R.M.A., no. 060353).

Table 2. Chemical analyses, $\text{Fe}^{2+}:\text{Mg}$ molecular ratios, optical data and specific gravity for 4 specimens of scorzalite together with lazulite from Hållsjöberget.

Locality	Corrego Frio	White Mts.	Victory Mine	Hållsjöberg* (scorzalite)	Hållsjöberg* (lazulite)
MgO	4.23	3.38	2.93	1.49	8.62
FeO	14.74	16.27	17.06	18.00	8.46
MnO	0.11	0.07	0.10	0.50	0.12
CaO	0.02	0.08	0.03	—	—
Al_2O_3	30.87	30.48	30.80	28.75	31.39
Fe_2O_3	0.54	0.24	0.13	3.03	0.04
TiO_2	0.23	0.32	0.10	—	—
P_2O_5	42.90	43.31	42.67	42.39	45.03
H_3O^+	5.86	5.46	6.10	5.76	6.20
Total	99.54	99.61	99.92	99.92	99.86
Molecular ratio $\text{Fe}^{2+}:\text{Mg}$	66 : 34	73 : 27	76 : 24	87 : 13	36 : 64
Index of re α	1.633	1.637	1.636	1.645	1.619
fraction β	1.663	1.667	1.666	1.674	1.649
$(\pm 0.002) \gamma$	1.673	1.677	1.676	1.680	1.655
Optical angle $2V$	$62^\circ(-)\text{calc.}$	$61^\circ(-)\text{calc.}$	$61^\circ(-)\text{calc.}$	$64^\circ(-) \pm 1^\circ$	$67^\circ(-) \pm 1^\circ$
Specific gravity	3.268	3.272	3.327	3.36	3.18

* Recalculated analyses.

Mineralogical description

Idiomorphic crystals of scorzalite have not been observed and the mineral occurs as irregular grains congregated in a fine-grained aggregate (maximum grain-size 2.5 mm). The colour of the mineral is very dark blue and thus differs from the somewhat lighter blue of lazulite. The scorzalite is furthermore strongly pleochroic with α colourless, β and γ azure. Cleavage seldom perfect, generally occurs along (110). Twinning commonly occurs on (101) and the twins are usually formed polysynthetically. The optical plane of the mineral is (010). Other optical constants, chemical analyses, $\text{Fe}^{2+}:\text{Mg}$ molecular ratios and the specific gravity, are given in Table 2, as well as earlier published scorzalite analyses.

The results indicate, that scorzalite from Hållsjöberget has higher indices of refraction, a greater optical angle and a higher specific gravity than previously described representatives of this mineral. This is probably mainly due to its high Fe^{2+} content (18.00 %). The analyses also show, that the Fe^{3+} content (3.03 %) is considerably higher than other examined specimens of scorzalite (max. 0.54 %). It does not as yet seem possible to establish the effect of the content on the optical properties.

X-ray data

The determinations of the d -values for scorzalite and lazulite were performed by means of a Seeman-Bohlin camera (Cr $K\alpha$ -radiation). Results given by Pecora and Fahey (1950) are included here for comparison.

Table 3.

Scorzalite (Hållsjöberget)		Scorzalite (Corrego Frio)		Lazulite (Hållsjöberget)	
<i>d</i>	Intensity	<i>d</i>	Intensity	<i>d</i>	Intensity
		6.17	m		
		4.72	m		
		3.60	w	3.60	1
				3.54	1
				3.50	1
				3.44	1
				3.37	1
		3.24	vs	3.24	9
		3.20	vs	3.20	10
		3.14	s	3.14	8
		3.08	m	3.08	8
2.54	10	2.55	m	2.55	7
2.35	2	2.34	vw	2.35	3
2.27	8	2.26	m	2.26	5
2.23	7	2.22	mw	2.22	5
2.12	6	2.05	vw	2.05	2
2.01	6	2.01	m	2.01	3
1.993	3	1.989	mw	1.986	3
1.978	6	1.973	m	1.978	5
				1.963	5
1.828	4	1.828	vw	1.821	4
1.814	3	1.811	mw	1.808	2
1.789	3	1.787	w	1.786	2
				1.746	1
				1.736	1
		1.682	vw	1.688	1
				1.677	1
				1.664	1
1.627	7	1.622	mw	1.620	5
1.607	4	1.601	w	1.602	4
1.586	2			1.581	2
1.578	3	1.574	(a group) m	1.573	3
1.573	2			1.568	3
				1.560	1
				1.547	1
1.542	6	1.541	mw	1.539	5
		1.412	w		
		1.390	vw		
		1.277	m		
		1.186	vw		
		1.115	vw		
		0.8318	vw		
		0.8294	vw		
		0.7849	vw		
		0.7746	vw		

As indicated in Table 3, the two minerals show only slight differences in the *d*-values, which has also been found in previous investigations (Pecora and Fahey, 1950).

ACKNOWLEDGEMENTS

The author wishes to convey his thanks to Persbergs Grufve AB for defraying the cost of the analyses, to Prof. F. E. Wickman, Riksmuseet, for his critical reading of the manuscript and to Mr L. Jansson, who carried out much of the laboratory work.

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Tryckt den 5 juni 1956

Uppsala 1956. Almqvist & Wiksells Boktryckeri AB

Magnussonite, a new arsenite mineral from the Långban mine in Sweden

By O. GABRIELSON

With a chemical analysis by R. BLIX

In the Långban mine a hitherto undescribed, green arsenite mineral occurs in association with fissure minerals. It is identical with mineral no. 42 in G. Flink's collection of new or incompletely described minerals from Långban (1923). According to a description by G. Flink (1920) of trigonite and dixenite from Långban, a green mineral with the same properties as the new arsenite mineral was found in the year 1919 accompanying dixenite on fissures in the dolomite in the "Hindenburg" ore.

The arsenite mineral is named after Professor N. H. Magnusson (born in 1890), Director of the Geological Survey of Sweden, who has made important contributions to our knowledge of the geology and mineralogy of the Långban mine.

Occurrence.—Magnussonite occurs as fine-grained incrustations in fissures, usually in hausmannite-impregnated dolomite or in fine-grained hematite together

Table 1.

	1		2	3
CuO	2.07	Cu	2.86	79.86
MnO	47.24	Mn	73.00	
MgO	1.47	Mg	4.00	
As ₂ O ₃	43.49	As	48.20	48
H ₂ O	1.16	OH	14.12	16.72
Cl	0.84	Cl	2.60	
		O	143.80	143.80
Insolubles (barite)	3.68			144
	99.95			
Less O for Cl	0.19			
	99.76			

1. Magnussonite, Långban (analyst R. Blix).
2. Number of atoms in the unit cell, density, 4.23, unit cell volume 4135 Å³.
3. Ideal number of atoms in the magnussonite unit cell.

Table 2. X-ray powder diffraction data of magnussonite, Långban, Sweden. Fe-radiation.

h k l	sin θ	d (obs)	d (calc)	I
2 0 0	0.121	8.01	8.03	1
3 0 0	0.184	5.26	5.35	< 1
2 2 1				
3 1 1	0.197	4.92	4.84	< 1
4 0 0	0.241	4.02	4.01	1
5 0 0	0.303	3.20	3.21	< 1
4 3 0				
5 1 1	0.311	3.12	3.09	3
3 3 3				
4 4 0	0.340	2.85	2.84	10
5 3 1	0.357	2.71	2.71	< 1
5 3 3	0.393	2.47	2.45	3
4 4 4	0.412	2.35	2.32	< 1
8 0 0	0.477	2.03	2.01	< 1
9 2 1	0.556	1.74	1.73	2
7 6 1				
6 5 5				
9 3 0	0.570	1.70	1.69	< 1
8 5 1				
7 5 4				
9 3 1	0.574	1.69	1.68	< 1
10 0 0	0.602	1.61	1.61	< 1
8 6 0				
10 3 3	0.653	1.48	1.48	1
9 6 1				
11 2 2	0.682	1.42	1.41	< 1
10 5 2				
8 8 1				
8 7 4				

with calcite, trigonite, dixenite and brown, manganiferous serpentine. Moreover, an unknown black mineral ($n=1.55$) with Fe, Mg, Cu and Si as chief constituents, is present according to a spectrographic determination.

Crystallographic and optical properties.—Magnussonite has not been found as separate crystals, but an optical investigation shows that magnussonite is isotropic and consequently has cubic symmetry. The refractive index is very high, $n=1.980 \pm 0.005$, determined by the immersion method.

Physical properties.—The color is grass-green to emerald-green, sometimes blue-green. Streak white. Luster vitreous. Hardness 3.5–4. The specific gravity was determined to be 4.30. The value of the specific gravity, calculated from the volume of the unit cell and the theoretical molecular weight, is 4.23.

Structural data.—The X-ray powder data of magnussonite, measured on a Philips Geiger counter X-ray spectrometer (Fe-radiation) are given in Table 2. From these data the edge of the cubic cell was calculated to be $16.05 \pm 0.05 \text{ \AA}$ and consequently the volume of the unit cell is 4135 \AA^3 .

The value of the cube edge will however not be definitely fixed before an investigation of the mineral structure has been performed. It is possible that some other length of the cube edge with a simple relationship to the value 16.05 Å is more suitable.

Chemical composition.—A chemical analysis of magnussonite has been performed by Dr. R. Blix. The result of this analysis is shown in Table 1, from which the unit cell content $(\text{Mn, Mg, Cu})_{80}\text{As}_{48}(\text{O, OH, Cl})_{160}$ was calculated, corresponding to sixteen molecules of $(\text{Mn, Mg, Cu})_5(\text{OH, Cl})(\text{AsO}_3)_3$. The formula of magnussonite is very similar to that of finnemanite, $\text{Pb}_5\text{Cl}(\text{AsO}_3)_3$, but the minerals are, of course, not isostructural, finnemanite having hexagonal symmetry and magnussonite cubic.

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Tryckt den 5 juni 1956

Uppsala 1956. Almqvist & Wiksells Boktryckeri AB

The crystal structure of thaumasite $\text{Ca}_3\text{H}_2(\text{CO}_3 / \text{SO}_4)\text{SiO}_4 \cdot 13 \text{H}_2\text{O}$

By ERIC WELIN

With 4 figures in the text

Previous investigations

Thaumasite was first described in 1878 by A. E. Nordenskiöld. In the same year the mineral was analysed by G. Lindström who proposed the formula $\text{CaCO}_3 \cdot \text{CaSO}_4 \cdot \text{CaSiO}_3 \cdot 14 \text{H}_2\text{O}$. According to O. Widman (1890) the formula should contain 15 water molecules. S. L. Penfield and J. H. Pratt (1896) showed that the mineral was hexagonal and E. T. Wherry (1917) determined it as holohedric with an axis ratio of $a:c = 1:0.931$. An X-ray investigation was published by G. Aminoff in 1933 who found thaumasite to belong to one of the space groups C6 , C6_3 , C6/m or $\text{C6}_3/\text{m}$. He found the dimensions of the unit cell to be $a = 10.90$ and $c = 10.29 \text{ \AA}$. Assuming a density of 1.914 and taking the number of water molecules as 15, which had already been proposed, he found the number of molecules contained in the unit cell to be two. Th. Vogt (1938) compared the structure of thaumasite on chemical evidence with that of beryl, which contains a closed ring of six SiO_4 tetrahedra of Z_6O_{18} type. H. Strunz (1949) placed thaumasite as a structurally unknown mineral in a subdivision of the olivine-norbergite group.

Material

A small acicular crystal (0.3–0.4 mm thick) from Långban, Sweden, showing prismatic faces, was used for the present investigation.

Cell dimensions and number of molecules in the unit cell

Rotation and equi-inclination photographs were taken around the c - and a -axis using unfiltered copper radiation. The cell dimensions were found to be $a = 10.95$ and $c = 10.30 \text{ \AA}$. Within the limits of error this is in accordance with Aminoff's data. A calculation of the number of molecules in the unit cell gave 1.96 when 15 water molecules and a density of 1.882 (Th. Vogt 1938) were used in the formula. With 14 water molecules the result was 2.02, when the same density was assumed. For this and other reasons, mentioned later, the formula, written according to Lindström's notation, should contain 14 water molecules.

Space group

As the cell contains two molecules we have to locate, in all, six calcium, two silicon, two sulphur, two carbon, twenty oxygen atoms and twenty-eight water

Crystal class	Space group	Reasons for exclusion
6/m	C _{6₃} /m C ₆ /m	Six-fold axis of rotary inversion in 2-fold point positions 001/4, 003/4; 1/3 2/3 1/4, 2/3 1/3 3/4 and 2/3 1/3 1/4, 1/3 2/3 3/4. Six-fold rotation axis in 2-fold position 00z, 00 \bar{z} . Six-fold axis of rotary inversion in 2-fold positions 1/3 2/3 0, 2/3 1/3 0 and 1/3 2/3 1/2, 2/3 1/3 1/2. No extinctions 000l.
6	C _{6₃} C _{6₄} C _{6₂} C _{6₅} C _{6₁} C ₆	None. Only 3- and 6-fold positions, reflections 000l if $l = 3n$. Only 3- and 6-fold positions, reflections 000l if $l = 3n$. Only 6-fold positions, reflections 000l if $l = 6n$. Only 6-fold positions, reflections 000l if $l = 6n$. Six-fold rotation axis in 1-fold point position 00z, only one 2-fold position 1/3 2/3 z, 2/3 1/3 z. No extinction 000l.
$\bar{6}$	C $\bar{6}$	No extinction 000l.

molecules in the cell. It is most probable that the sulphur atoms together with oxygen atoms appear as tetrahedric SO₄ groups and the carbon atoms as plane CO₃ groups. The silicon atoms can appear either as one Si₂O₇ group or as two SiO₄ groups. This latter configuration seems to be the more probable one.

In the recorded photographs no reflections from 000l could be observed in odd orders. As the Laue symmetry is 6/m, thaumasite belongs to one of the crystal classes 6/m, 6 or $\bar{6}$. By taking into account the symmetry of the atomic groups assumed, the symmetry of the point positions as well as the observed extinctions, the space groups of the crystal classes mentioned have been examined in order to find the correct space group. This is summarized in the table above.

The only space group left is C_{6₃} and it was thus taken as basis for a trial structure.

Positions of the atoms

The structure was determined by means of the trial-and-error method. The point positions of C₆⁶-C_{6₃} are

- 2: (a) 00z, 001/2 + z (b) 1/3 2/3 z, 2/3 1/3 1/2 + z
6: (c) x y z, \bar{y} x-y z, y-x \bar{x} z, \bar{x} \bar{y} 1/2 + z, y y-x 1/2 + z, x-y x 1/2 + z

In the point position (a) the distance between two atoms of the same kind has to be 1/2 + z which in this case means 5.15 Å. For this reason it is impossible to locate one Si₂O₇ group, with a distance between the Si atoms of 3.2 Å, in position (a) which is on the screw axis 6₃. In case (b) the two positions are on different three-fold rotation axes, which fact also prohibits the location of one Si₂O₇ group in these points. Thus the structure must contain SiO₄ tetrahedra which together with SO₄ and CO₃ groups occupy the positions (a) and (b). If the SiO₄ tetrahedra are supposed to be in the point position (a) they will also be on the axis 6₃ and have one face of the tetrahedron perpendicular to this axis, the tetrahedron corners on the axis all pointing in the same direction. The length of c is approximately twice the height of a SiO₄ tetrahedron.

If the SiO₄ tetrahedra occupy the position assumed above the only position left for the SO₄ and CO₃ groups is (b). Further discussions on this assumption will be

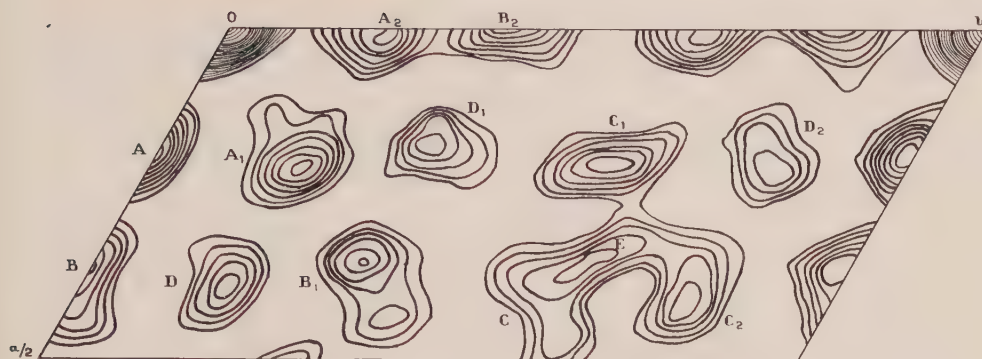


Fig. 1. Patterson projection of thaumasite on (0001). Contour lines are drawn at height differences of 100 units.

found on page 142. Since the rotation axis in (b), as already mentioned, is trigonal, the SO_4 tetrahedron has to be located like the SiO_4 one, i.e. with one face perpendicular to the rotation axis. From a structural as well as from a chemical point of view the apices of the SO_4 tetrahedra could be placed equally well in the same as in the opposite direction of the SiO_4 tetrahedra. By comparing the calculated and observed intensities, a slightly better agreement was obtained in the latter case.

The mineral is optically negative and the CO_3 group has to lie in a plane perpendicular to the rotation axis. If a water molecule is placed in position (b) between the CO_3 and SO_4 groups all of the available space on this axis is occupied.

According to the Patterson projection on (0001) (Fig. 1) the mineral is highly symmetrical along the axis a and b . The SiO_4 , SO_4 and CO_3 groups in the 2-fold positions (a) and (b) do not give rise to that symmetry distribution. However, with one or more 6-fold point positions on the axis the symmetry in the projection could be explained. If the highest maximum A (A_1 , A_2) is ascribed to the vectors Ca–Ca, Ca–Si and Ca–OI a chain SiO_4 –Ca– SiO_4 is formed and the Ca parameters are found to be $x = 0$, $y \approx 0.2$. The distance between two calcium atoms around the same SiO_4 tetrahedron is in this case about 3.9 Å, which is the normal value. This Ca–Ca vector is represented by the maximum D (D_1 , D_2). The maximum C (C_1 , C_2) is probably built up by almost coinciding vectors from Ca atoms to S, C, H_2OI , H_2OII and confirms the supposed Ca parameters. These parameters are also verified by the peak B (B_1 , B_2), which represents the vector Ca–OII.

The remaining twenty-four water molecules occupy the general position (c). As a result three of these four 6-fold groups together with the oxygen atoms from SiO_4 form an almost octahedric six coordination around every Ca atom.

A projection of this structure on (0001) shows that there is an apparent centre of symmetry at $x = 1/2$ and $y = 1/2$. Hence there is no difficulty in performing a Fourier synthesis for a more accurate determination of the x and y parameters. Such syntheses were carried out with $|F|$ -values calculated from the visually estimated intensities in the Weissenberg photographs in order to improve the accuracy of the x and y parameters. No corrections for absorption were made. The $|F|$ -values for the hkl and $hk2$ reflections were calculated from the estimated intensities by the method of Lu (1943).

The z parameters were determined geometrically from the more carefully deter-

Table 1.

Final parameters				
6 Ca	in (c)	$x=0$	$y=0.210$	$z=0.295$
2 Si	„ (a)	$x=0$	$y=0$	$z=0$
2 C	„ (b)	$x=1/3$	$y=2/3$	$z=0.530$
2 S	„ (b)	$x=1/3$	$y=2/3$	$z=0.955$
2 O I	„ (a)	$x=0$	$y=0$	$z=0.165$
6 O II	„ (c)	$x=0$	$y=0.115$	$z=0.900$
6 O III	„ (c)	$x=0.195$	$y=0.605$	$z=0.030$
6 O IV	„ (c)	$x=0.195$	$y=0.605$	$z=0.530$
2 H ₂ O I	„ (b)	$x=1/3$	$y=2/3$	$z=0.810$
2 H ₂ O II	„ (b)	$x=1/3$	$y=2/3$	$z=0.280$
6 H ₂ O III	„ (c)	$x=0$	$y=0.355$	$z=0.465$
6 H ₂ O IV	„ (c)	$x=0$	$y=0.465$	$z=0.810$
6 H ₂ O V	„ (c)	$x=0.230$	$y=0.390$	$z=0.190$
6 H ₂ O VI	„ (c)	$x=0.400$	$y=0.210$	$z=0.170$

Table 2. Weissenberg photograph of thaumasite. Rotation axis [0001]. Zero layer line. Cu-K α radiation. Comparison between observed and calculated intensities.

	0 <i>k</i> 0	1 <i>k</i> 0	2 <i>k</i> 0	3 <i>k</i> 0	4 <i>k</i> 0	5 <i>k</i> 0	6 <i>k</i> 0	7 <i>k</i> 0	8 <i>k</i> 0	9 <i>k</i> 0
	obs calc	obs calc	obs calc	obs calc	obs calc	obs calc	obs calc	obs calc	obs calc	obs calc
<i>h</i> 9 0	16 18	7 4								
<i>h</i> 8 0	5 5	9 10	6 4							
<i>h</i> 7 0	3 3	5 5	3 1	2 3	3 4					
<i>h</i> 6 0	21 15	5 1	<1 0	8 15	<1 2	1 1				
<i>h</i> 5 0	33 33	7 3	<1 1	<1 1	3 5	12 18				
<i>h</i> 4 0	3 2	9 13	5 7	8 7	2 13	6 4	2 2	7 8		
<i>h</i> 3 0	13 13	3 4	11 10	5 8	5 3	5 3	22 16	2 6		
<i>h</i> 2 0	5 5	4 4	7 8	7 7	<1 0	2 5	2 3	<1 2	3 6	
<i>h</i> 1 0	10 9	13 20	1 2	7 4	3 9	2 1	2 3	7 9	3 7	4 5

Table 3. Weissenberg photograph of thaumasite. Rotation axis [0001]. First layer line. Cu-K α radiation. Comparison between observed and calculated intensities.

	0 <i>k</i> 1	1 <i>k</i> 1	2 <i>k</i> 1	3 <i>k</i> 1	4 <i>k</i> 1	5 <i>k</i> 1	6 <i>k</i> 1	7 <i>k</i> 1	8 <i>k</i> 1
	obs calc	obs calc	obs calc	obs calc	obs calc	obs calc	obs calc	obs calc	obs calc
<i>h</i> 8 1	4 2	4 6							
<i>h</i> 7 1	2 2	6 4	6 4	4 7					
<i>h</i> 6 1	<1 2	2 5	12 10	8 4					
<i>h</i> 5 1	3 3	3 4	3 3	4 5	3 2	2 1			
<i>h</i> 4 1	<1 3	3 4	5 10	3 3	5 5	3 3	3 4		
<i>h</i> 3 1	2 1	5 6	4 4	13 17	8 8	3 2	4 5		
<i>h</i> 2 1	3 3	10 14	2 4	12 6	7 6	3 5	8 9	5 4	4 3
<i>h</i> 1 1	2 2	5 10	6 11	7 4	3 4	6 6	4 9	8 5	4 4

Table 4. Weissenberg photograph of thaumasite. Rotation axis [0001]. Second layer line. Cu-K α radiation. Comparison between observed and calculated intensities.

	0 <i>k</i> 2 obs calc	1 <i>k</i> 2 obs calc	2 <i>k</i> 2 obs calc	3 <i>k</i> 2 obs calc	4 <i>k</i> 2 obs calc	5 <i>k</i> 2 obs calc	6 <i>k</i> 2 obs calc	7 <i>k</i> 2 obs calc	8 <i>k</i> 2 obs calc
<i>h</i> 9 2	7 11								
<i>h</i> 8 2	<1 2	6 6	2 5						
<i>h</i> 7 2	3 4	8 7	5 3	8 5					
<i>h</i> 6 2	8 8	8 9	8 7	10 5	4 5				
<i>h</i> 5 2	7 6	7 6	5 5	4 5	<1 6	3 1			
<i>h</i> 4 2	10 15	10 11	6 8	6 9	8 4	9 8	9 7		
<i>h</i> 3 2	17 13	11 13	2 9	12 11	6 9	6 4	7 10	2 4	
<i>h</i> 2 2	8 9	<1 12	3 7	10 9	1 5	8 6	5 5	3 8	11 6
<i>h</i> 1 2	10 11	13 13	3 14	5 10	4 7	6 3	7 7	13 8	3 3

Table 5. Weissenberg photograph of thaumasite. Rotation axis [0110]. Zero layer line. Cu-K α radiation. Comparison between observed and calculated intensities.

	0 0 <i>l</i> obs calc	1 0 <i>l</i> obs calc	2 0 <i>l</i> obs calc	3 0 <i>l</i> obs calc	4 0 <i>l</i> obs calc	5 0 <i>l</i> obs calc	6 0 <i>l</i> obs calc	7 0 <i>l</i> obs calc	8 0 <i>l</i> obs calc	9 0 <i>l</i> obs calc	10 0 <i>l</i> obs calc
<i>h</i> 0 12	8 10										
<i>h</i> 0 11	— —										
<i>h</i> 0 10	9 12		7 4	2 3	2 6	8 8		6 3			
<i>h</i> 0 9	— —	2 4	<1 5	2 3	<1 3	<1 2		<1 2	3 2		
<i>h</i> 0 8	16 22	11 5	<1 5	5 5	6 5	11 15	6 3	3 5	4 8	6 7	
<i>h</i> 0 7	— —	4 4	3 5	<1 4	6 4	<1 4	<1 1	5 4	<1 3	2 1	
<i>h</i> 0 6	28 22	11 4	4 2	5 8	<1 8	11 13	5 13	<1 5	<1 1	3 12	5 5
<i>h</i> 0 5	— —	7 1	4 4	4 2	2 3	5 2	3 2	4 2	5 2	<1 1	<1 2
<i>h</i> 0 4	23 20	2 5	8 6	<1 9	6 11	17 15	5 6	4 2	5 3	3 7	10 8
<i>h</i> 0 3	— —	4 5	2 13	<1 1	5 3	2 3	<1 2	<1 3	<1 3	4 1	<1 2
<i>h</i> 0 2	5 11	10 11	8 9	17 13	10 15	7 6	8 8	3 4	<1 2	7 11	6 2
<i>h</i> 0 1	— —	2 2	3 3	2 1	<1 3	3 3	<1 2	2 2	4 2	<1 2	<1 2
<i>h</i> 0 0		10 9	5 5	13 13	3 2	33 33	21 15	3 3	5 5	16 18	7 14

mined x and y parameters and checked against the observed intensities. The accuracy of the x and y parameters has been estimated to be ± 0.01 , while the accuracy of the z parameters is probably less.

The final parameters with one of the silicon atoms arbitrarily chosen as origin are given in Table 1.

The comparison between observed and calculated intensities is shown in the Tables 2–5.

Description of the structure

The structure of thaumasite is shown projected on (0001) in Fig. 2. This projection and Fig. 3, which is a drawing of a part of the unit cell, show each sulphur and each silicon atom in the middle of an assumed tetrahedron formed by oxygen atoms. The distances S–O used in the SO₄ tetrahedron, 1.51 Å, and Si–O in the SiO₄ tetrahedron,

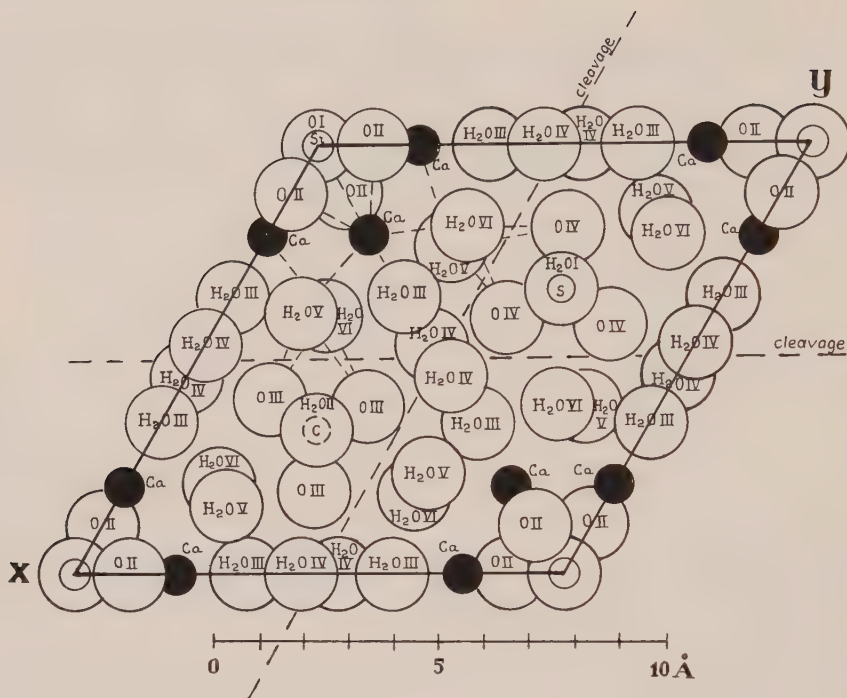


Fig. 2. The structure of thaumasite projected on (0001).

1.61 and 1.69 Å, are the normal distances found in other structures. The normal values are also chosen for the carbon atoms, i.e. they are surrounded by three oxygen atoms with interbond angles of 120° and three equal C–O bonds of length 1.31 Å.

In the structure examined, the question whether the sulphur or the silicon occupy position (a) could not be decided by intensity calculations as their scattering powers are almost equal. The carbon atom, which can also come into question, has on the contrary a lower scattering power and can be assumed excluded from occupying that position. Probably the silicon atoms occupy position (a) while a chain formation for Ca–SiO₄ seems to be more likely, the electrostatic attraction between Ca–SiO₄ in addition being stronger than between Ca–SO₄. In the case of a bond Ca–SiO₄ a chain along the *c*-axis is formed. Three water molecules, which together with three silicon-bonded oxygens form a slightly distorted octahedron around the calcium atom, are on their other side connected with hydrogen bonds to the SO₄ and CO₃ groups respectively. Consequently one of those water molecules is bound to a calcium and to two oxygen atoms in a SO₄ or CO₃ group.

The SO₄ and CO₃ groups are similar structural units in the structure under consideration. The difference in electrostatic valency between the oxygen ions in the respective groups is offset by two hydrogen atoms that appear most likely in connection with the SO₄ group.

Owing to the fact that the cations are wholly enclosed by oxygen atoms and water

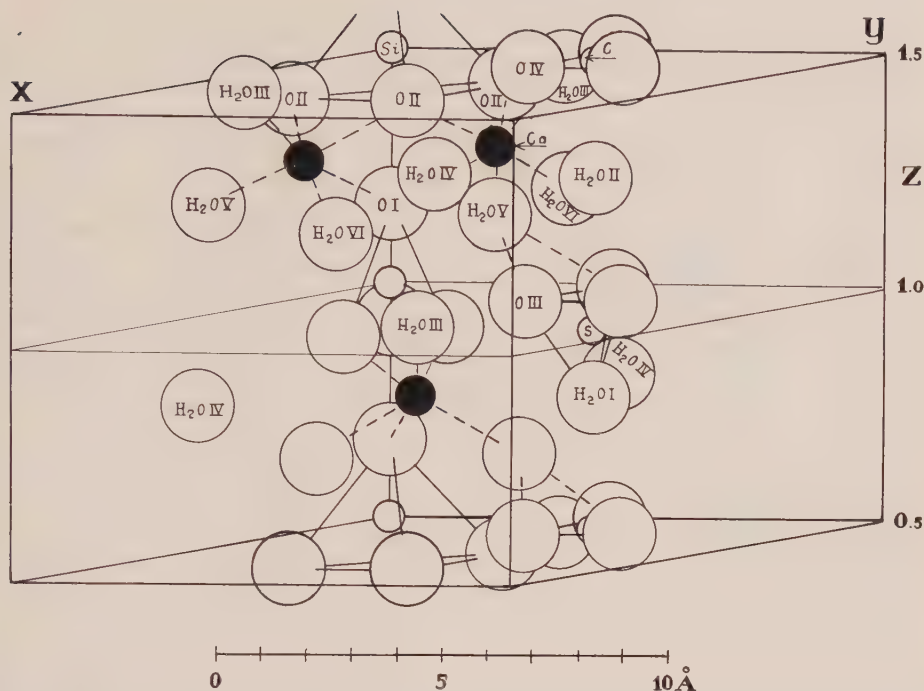


Fig. 3. Detail of the structure of thaumasite. The cell is displaced one half unit.

molecules the fourth water molecule in the 6-fold position is surrounded only by anions and water molecules.

In a review of crystalline salt hydrates Jensen (1948) summed up the characteristic appearance of the water molecules. These rules have been criticised and found to be wrong in part by Lindqvist (1948) and Dahlman (1952). The appearance of the water molecules in the structure of thaumasite, however, does not contradict the rules of Jensen. Three of the four 6-fold water positions are coordinated with one cation (Ca) and the other side with two anions. These atoms lie almost in the same plane and form an inequilateral triangle around the water molecule, which is not exactly in the middle of the triangle. As already stated the water molecules in the fourth 6-fold position and the 2-fold position are not in contact with any cations since the latter are already wholly surrounded by other ions and molecules. Whether the last-mentioned 6-fold water molecules are bound to their nearest neighbours, i.e. oxygen ions and water molecules, can only be decided by accurate determinations of the positions and the distances between these atoms.

In the introduction it was pointed out that two formulas for the thaumasite have been proposed, one with 14 H_2O and the other with 15 H_2O . However, the 2- and 6-fold point positions and the space available in C6_3 exclude the formula with 15 H_2O . The calculations of the molar refractivity (Holden 1922) and the number of molecules in the unit cell also show the formula proposed by Widman to be incorrect. No investigation with the water molecules eliminated has been undertaken as it seems impossible to remove the water without breaking down the structure.

Table 6. The distances between the atoms and their nearest neighbours.

Kind of neighbour		Number	Distance in Å
Ca	O I, O II, H ₂ O III,	1, 2, 1	2.66, 2.28, 2.35
	H ₂ O V, H ₂ O VI	1, 1	2.55, 2.60
Si	O I, O II	1, 3	1.69, 1.61
C	O IV, H ₂ O I, H ₂ O II	3, 1, 1	1.31, 2.89, 2.58
S	O III, H ₂ O I, H ₂ O II	3, 1, 1	1.51, 1.51, 3.33
O I	O II, H ₂ O V, H ₂ O VI	3, 3, 3	2.75, 3.71, 3.78
O II	H ₂ O III, H ₂ O IV, H ₂ O V	2, 1, 1	3.46 3.94, 4.07
	H ₂ O VI	1	3.92
O III	H ₂ O I, H ₂ O II, H ₂ O III	1, 1, 1	2.60, 2.89, 2.48
	H ₂ O IV	2	3.45 and 2.97
	H ₂ O V	2	2.60 and 2.98
O IV	H ₂ O I, H ₂ O II, H ₂ O III	1, 1, 1	3.17, 2.89, 2.61
	H ₂ O IV	2	3.47 and 2.95
	H ₂ O VI	2	2.61 and 2.67
H ₂ O I	H ₂ O VI	3	2.94
H ₂ O II	H ₂ O V	3	2.83
H ₂ O III	H ₂ O IV, H ₂ O V	2, 1	2.57 and 3.76, 2.78
	H ₂ O VI	1	2.83
H ₂ O IV	H ₂ O V	2	2.58 and 4.93
	H ₂ O VI	2	2.88 and 4.59
H ₂ O V	H ₂ O VI	1	3.42

Thaumasite usually grows as prismatic crystals, elongated parallel to the *c*-axis. It has been pointed out that the crystals are built up of chains of $-\text{Ca}-\text{SiO}_4-\text{Ca}-$ stretching indefinitely in the direction of the *c*-axis. The forces in this direction are stronger than those between the individual chains and the CO_3 and SO_4 groups. This structural feature explains the tendency of the crystal to be elongated along the *c*-axis. It also explains the observed prismatic cleavage parallel to (1010). This zone of weakness in the crystal is shown by dashed lines in Fig. 2.

All the facts mentioned give us the key for writing the chemical formula of thaumasite. The most important part of the structure, the $\text{Ca}-\text{SiO}_4$ chain must be clearly expressed in the formula. The function of the SO_4-CO_3 groups as similar structural units, the important role of the oxygen atoms as connectors between the $\text{Ca}-\text{SiO}_4$ chain and SO_4/CO_3 groups as well as the fact that hydrogen atoms take part in this connection have to be indicated in the formula. In order to concentrate all these chemical and structural properties of the groups the formula of thaumasite has been written $\text{Ca}_3\text{H}_2(\text{SO}_4/\text{CO}_3)\text{SiO}_4 \cdot 13\text{H}_2\text{O}$.

The distances between the atoms and their nearest neighbours are given in Table 6.

Relation of other structures to thaumasite

Although no other compounds with SiO_4 , CO_3 and SO_4 or structurally comparable radicals are known a comparison might be attempted with unhydrated or hydrated compounds of calcium and of any of the radicals mentioned above.

The structures of the following calcium orthosilicates are known: $\alpha\text{-C}_2\text{S}$, $\alpha'\text{-C}_2\text{S}$, $\beta\text{-C}_2\text{S}$, $\gamma\text{-C}_2\text{S}$ and C_3S .* Of these compounds only $\gamma\text{-C}_2\text{S}$ shows structural features

* $\text{C} = \text{CaO}$, $\text{S} = \text{SiO}_2$, $\text{H} = \text{H}_2\text{O}$.

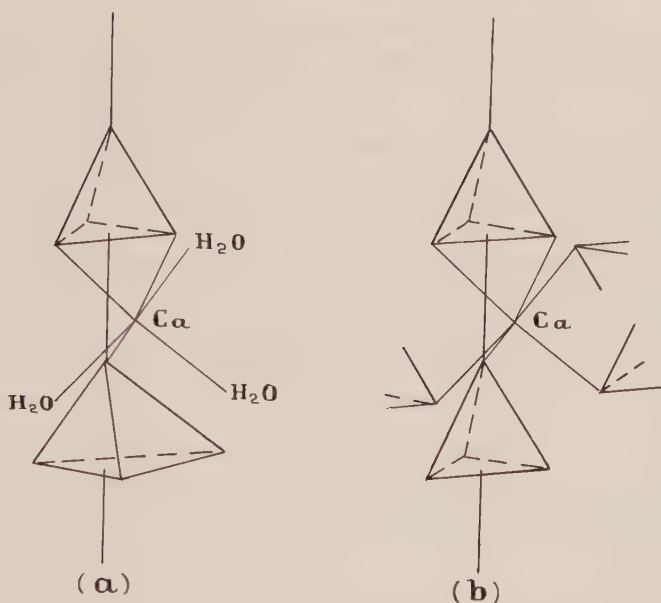


Fig. 4. Comparison between the Ca-SiO₄ chains in thaumasite (a) and in γ -C₂S (b).

comparable to thaumasite. According to O'Daniel and Tscheischwili (1942) γ -C₂S has an olivine structure and belongs therefore to the space group Pbnm. The calcium atoms in this structural type occupy two different positions. CaI atoms are located in the symmetry centra and the CaII atoms on the mirror planes. In monticellite the first group is replaced by magnesium. In both cases the calcium atoms are six coordinated and the surrounding oxygen atoms form a slightly distorted octahedron. CaI is bound to two oxygens in two different tetrahedra and to one oxygen in two other tetrahedra. The CaII atoms are bound to two oxygens in one tetrahedron and to four oxygens in four different tetrahedra. This configuration can be compared with the Ca-SiO₄ chain in thaumasite, where three of the four oxygen atoms from the CaII coordination in γ -C₂S are replaced by water molecules. This comparison is shown in Fig. 4.

The remaining calcium orthosilicates do not show the same pronounced similarities with the Ca-SiO₄ chain in thaumasite as does γ -C₂S. Their space groups, structural types and calcium coordination are

α -C ₂ S	C $\bar{3}$ m	glaserite	6, 10	Bredig (1942)
α' -C ₂ S	Pmcn	K ₂ SO ₄	9, 10	Bredig (1942)
β -C ₂ S	P2 ₁ /n		6 + 6, 8	Midgley (1952)
γ -C ₂ S	Pbnm	olivine	6,	O'Daniel & Tscheischwili (1942)
C ₃ S	R3m		6	Jeffery (1952)

Six coordination, which also appears in thaumasite, is common. The mean distances Ca-O are in β -C₂S 2.47 Å and 2.49 Å for the two different Ca positions and in C₃S, 2.40 Å. In accordance with Pauling's rules it is also clear from the structures mentioned that calcium is usually bound to one oxygen ion in different silicon tetrahedra,

sometimes to two oxygen ions in the same tetrahedron and more rarely to three oxygen ions in one tetrahedron.

The structures of the hydrated calcium orthosilicates are known in a few cases only. The best investigated of those is awillite, $\text{Ca}_3(\text{SiO}_3\text{OH})_2 \cdot 2\text{H}_2\text{O}$ (Megaw 1952). The structure of that mineral is built from isolated silicon tetrahedra, linked together through edges and corners by calcium polyhedra to form a fairly closely packed arrangement. In each silicon tetrahedra there is an hydroxyl instead of an oxygen in one of the corners. The steric calcium coordination is seven, the electrostatic coordination being six. One water molecule is located in an approximately tetrahedral configuration; the other one in a plane triangle. The method of linking together the silicon tetrahedra is not the same in thaumasite as in awillite. A comparison of the interatomic distances between the oxygen ions of the two minerals is of importance if it can show that bonds of the same kind as those found in awillite also exist in thaumasite. In the structure of this mineral, O-O distances less than 2.80 Å (2.48–2.67 Å) have to be assumed between oxygen atoms of the SO_4/CO_3 groups and oxygens of the calcium polyhedra. Distances from the sixth water molecule to surrounding oxygen atoms are also short (2.57–2.58 Å). In awillite O-O distances corresponding to short and long hydrogen bonds are 2.523 Å and 2.716 Å respectively. The short O-O distances in thaumasite may consequently represent both kinds of hydrogen bonds and it is reasonable to assume that the bond lengths, 2.48–2.60 Å, are short hydrogen bonds while those exceeding 2.60 Å are long ones.

Jeffery (1951), discussing $\alpha\text{-C}_2\text{SH}$, states that the analysis indicates calcium ions, SiO_4 groups and oxygen atoms. It appears probable that one hydrogen atom is attached to the SiO_4 group and the other to the oxygen atom. In regard to the $\beta\text{-C}_2\text{SH}$ (hillebrandite) he says that if the measured spacing 3.63 Å (*a*-axis) is right, the SiO_4 tetrahedra can only be packed in a way that almost certainly require hydrogen binding between them. Consequently in both minerals it seems as if the OH groups are directly linked to the silicon atoms as they are in awillite, Taylor (1950) points out that CSH I probably has a layer structure in which individual distorted calcium hydroxide layers are relatively well crystallised. This layer structure is similar to those of the clay minerals as far as the variable spacing between the layers is concerned, but differs from them as regards the structure of the layers. It is evident that too little is known of the structures of these hydrated calcium silicates to make a comparison with thaumasite possible.

Recently an investigation of tilleyite, $\text{Ca}_5\text{Si}_2\text{O}_7(\text{CO}_3)_2$, has been published (Smith 1953). The mineral is so far the only structurally known silicate with CO_3 groups. A comparison with the structure of thaumasite may therefore be of interest.

The crystals of tilleyite are monoclinic and the space group is $\text{P}2_1/\text{a}$. According to the author the structure "is best regarded as an arrangement of isolated Si_2O_7 - and CO_3 -groups linked together by calcium atoms and held apart by a few oxygen atoms of different groups". The calcium atoms in this structure occupy five different positions. In three of them the calcium atoms are six-coordinated, in the two other positions the steric coordination is seven but the electrostatic is five. The number of Si_2O_7 and CO_3 groups linked to each calcium atom varies between four and six. Further "the Ca-O bonds are nearly parallel to one of three directions, one in the (204) plane and the two others symmetrically inclined to this plane. The Ca-O distances vary from 2.17 Å to 2.93 Å, the mean value being 2.40 Å." In the CO_3 group the carbon atom is asymmetric and one O-O distance is less than the two other, which are equal. The distances C-O are 1.33 Å which is somewhat larger than normal.

It is evident that the structures of the minerals are not very similar. Octahedric coordination occurs in both minerals. The SiO_4 groups in the thaumasite are however, linked to the CO_3 and SO_4 groups respectively through both calcium atoms and water molecules.

In gypsum, $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, calcium is bound to six SO_4 oxygens and two water molecules (Wooster 1936). The oxygen atoms belong to four different sulphate groups, of which two groups contribute two oxygens each. Besides being bound to calcium, the water molecules are also bound to two oxygens from different sulphate groups. In thaumasite there are also water molecules bound to calcium but on their other side to two oxygens in one sulphate group. The $\text{Ca}-\text{H}_2\text{O}$ distance is 2.44 Å in gypsum as compared to the mean value of 2.50 Å in thaumasite. The average distances from the water molecules in calcium polyhedra to the oxygen atoms of the SO_4 group in four different sulphates of the type $\text{M}^{\text{II}}(\text{SO}_4) \cdot \text{XH}_2\text{O}$ are 2.70 Å.

ACKNOWLEDGMENTS

This investigation has been carried out at the Mineralogical Department of the Swedish Museum of Natural History. The collections of minerals and the equipment were kindly placed at my disposal by Professor Frans E. Wickman to whom my thanks are due also for his interest and helpful discussions.

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Tryckt den 4 september 1956

Uppsala 1956. Almqvist & Wiksells Boktryckeri AB

An iron-rich wagnerite, formerly named talktriplite, from Hållsjöberget (Horrsjöberget), Sweden

By ÅKE HENRIQUES

Abstract

Talktriplite, originally described from Hållsjöberget by L. J. Igelström (1882) is an iron-rich wagnerite. New chemical analysis, X-ray data, the specific gravity and optical properties of the mineral are given.

Introduction

In 1882 L. J. Igelström described a new phosphate from Hållsjöberget (Horrsjöberget) in Värmland. Because of the chemical similarity of the mineral to triplite, from which it differs principally by having an extremely high magnesium richness, Igelström called the mineral talktriplite. His analysis was carried out on a very impure material and no quantitative determination of the fluorine content was done. As neither an X-ray examination nor an optical determination has been carried out since, the author is of the opinion that he may be justified in describing the mineral anew.

A powder photogram soon showed that the talktriplite was not a triplite variant, but instead a wagnerite mineral. The mineral differs from previously analysed wagnerite minerals mainly through a comparatively high content of iron and manganese. Considering this fact the mineral ought to be named iron-wagnerite.

Only the *d* values are given here, but single crystal work will be carried out in the near future.

The quartzite at Hållsjöberget

The kyanite-quartzite occurrence at Hållsjöberget is comparatively small. It belongs to the Pre-Cambrian basement and is interbedded in salic gneisses and grey pseudo-porphyrific rocks. In addition hyperites have fractured the quartzite which is frequently banded with alternating bands rich in kyanite and quartz respectively. In addition to these minerals the quartzite as a rule also contains rutile, lazulite, scorzalite and micaceous minerals. Further, small amounts of pyrite, haematite, ilmenite, garnet, tourmaline and pyrophyllite also occur. In addition to these minerals a number of other phosphate minerals, besides those mentioned above, are found within the quartzite, apatite, amphithalite, tetragophosphite, svanbergite and several other not previously described.

Investigation and treatment of the material

The material for chemical analysis and the other determinations was selected from a sample belonging to the collections of the Swedish Museum of Natural History, which was collected by L. J. Igelström. The richness of wagnerite in this sample has been estimated to 2–3 %. The purifying of the material has been carried out by separation by using heavy liquids on the sieved material which had a granular size of + 100 mesh (Tyler standard). As the material remained incompletely purified through this method a further separation was carried out by means of an isodynamical magnetic separator. Finally the material was hand-picked under the microscope. In spite of the above precautions the homogeneity of the material appeared incomplete (see below).

The refractive determinations were carried out by the immersion method. Other optical determinations were performed with Universal Stage as well as the cleavage indices. The optical angle was measured from sections in which the emergents of both optical axes could be seen through direct rotation from one optical axis to the other. Sodium-light was used for all determinations. The specific gravity was measured by the suspension method with the assistance of homogeneous grains (+ 50 mesh), temp. 20°C.

Chemical properties

The chemical composition of wagnerite is shown in Table 1. The primary chemical analysis has been converted to 100 % after discounting an insoluble residue of 2.23 % Al_2O_3 , Fe_2O_3 and TiO_2 , and 1.23 % SiO_2 . It is seen from the analysis that the wagnerite from Hållsjöberget differs from previously analysed wagnerites through having a considerably greater richness of FeO and MnO, 13.84 % and 8.00 % respectively, as earlier a maximum of 4.32 % FeO was given by Richmond (1940) and 0.63 % MnO by Hegemann & Steinmetz (1927). In addition the wagnerite from Hållsjöberget contains a comparatively high CaO content, 3.94 %. High CaO content has principally been found in wagnerite minerals altered to a greater or lesser degree. Thus Hegemann & Steinmetz (1927) showed a maximum CaO content of 13.45 % in strongly altered crystals from Werfen. The products of alteration consist in the main of apatite (Pisani 1897 and Hegemann & Steinmetz 1927). In Hållsjöberget the wagnerite is often found in conjunction with apatite of low manganese content which appears partly as an alteration product of wagnerite and partly in a more concentrated form which is not directly connected with the wagnerite mineral. It has not been possible to specify the exact proportion between the amount of CaO that will be part of the wagnerite itself and the amount that is found in apatite and eventually in other phosphates (the former is probably the lesser). The powder photograms show no lines which might be derived from apatite.

Mineralogical description

The physical properties of the mineral are shown in Table 2. For comparison a wagnerite from Werfen (MgO 48.33 %, FeO 0.95 %, P_2O_5 43.43 %, F 11.48 % and Rem. 0.70 %) described by Hegemann & Steinmetz (1927) has been included in the table. A comparison between the optical data of the two minerals shows great

Table 1. Chemical analyses of "talktriplite".

	1	2	3	4
CaO	3.81	3.94	0.0703	14.91
MnO	7.72	8.00	0.1128	14.86
FeO	13.36	13.84	0.1926	16.12
MgO	27.93	28.93	0.7175	17.42
P ₂ O ₅	37.38	38.72	0.2728	32.82
F	6.56	6.80	0.3579	n. d.
H ₂ O > 110°	2.54	2.63	0.1459	—
Al ₂ O ₃ , Fe ₂ O ₃ & TiO ₂	2.23		0.5038 (OH)	
SiO ₂	1.23			
	102.76 %	102.86 %		
Less O for F	2.76	2.86		
	100.00 %	100.00 %		

- 1) Wagnerite from Hållsjöberget (Analyst A. Parvel).
- 2) Recalculated analysis.
- 3) Molecular proportions.
- 4) "Talktriplite" from Hållsjöberget (Analyst L. J. Igelström).

The molecular proportions give the following ratio: R (Ca, Mn, Fe, Mg):PO₄:(F, OH) = 2.01:1.00:0.93, corresponding well with the accepted formula of wagnerite R₂PO₄F. R is approximately Mg_{0.66}Fe_{0.18}Mn_{0.10}Ca_{0.06}. Twenty-nine per cent of the fluorine is replaced by hydroxyl.

dissimilarities. The refractive indices as well as the optical angle show a striking increase in the wagnerite from Hållsjöberget which might be connected with the increased iron-manganese content. Differing from the wagnerites free from iron or with a low iron content the mineral now under examination shows a pronounced yellow colour in transmitted light. With regard to the refractive indices of triplite Heinrich (1951) found that (1) the refractive indices increase with the amount of FeO, (2) they decrease with the MgO and CaO content, (3) the increase per unit of FeO is considerably greater than the decrease per unit of MgO and CaO, and (4) the MnO content does not influence the refractive indices to any great extent. It does not seem unlikely that this will also prove to be the case with wagnerite, as future investigations may show. Other data in Table 2 should not require any further comment.

Paragenesis

The wagnerite occurs in low concentrations (max. about 3%) together with phosphate minerals such as lazulite, scorzalite, apatite etc., and quartz, rutile, ilmenite and muscovite within generally kyanite-rich parts of the quartzite. The high content of fluorine in the mineral has no counterpart in the other quartzite minerals with exception of rarely occurring phosphates (apatite etc.) and turmaline, and is probably due to a later limited supply of fluorine. This has probably occurred in connection with the formation of several narrow quartz–muscovite–pyrophyllite-rich zones within the quartzite. That is, the wagnerite is mainly found in connection

Table 2. The physical properties of wagnerite from Hållsjöberget and from Werfen.

	Hållsjöberget	Werfen
Habit	Massive (grains < 3 mm)	Many forms observed
Cleavage	Imperfect along $\{100\}$, $\{120\}$ and $\{010\}$	
Hardness	Ca. 5.5	
Lustre	Vitreous to resinous	
Colour	Yellow-red to brick-brown	
Colour in thin section	X = colourless-pale yellow Y = intense yellow Z = intense yellow X < Y < Z	
Index of refraction	$\alpha = 1.608 \pm 0.002$ $\beta = 1.615 \pm 0.002$ $\gamma = 1.630 \pm 0.002$	1.5678 1.5719 1.5824
Optical angle	$66^\circ (+) \mp 1^\circ$	$28^\circ 24 \frac{1}{2}' (+)$
Orientation	Y = b Z \wedge c = $53^\circ \mp 5^\circ$	Y = b Z \wedge c = $-21 \frac{1}{2}^\circ$
Specific gravity	3.47	

Table 3. X-ray data

The determinations of the d -values for wagnerite were carried out with the aid of a Guinier camera (Cu $K\alpha$ -radiation). For comparison the d -values for wagnerite from Bamle, Norway, have been included.

Wagnerite (Hållsjöberget)		Wagnerite (Bamle)		Wagnerite (Hållsjöberget)		Wagnerite (Bamle)	
d Å	Intensity	d Å	Intensity	d Å	Intensity	d Å	Intensity
		5.635	2	1.929	3	1.915	1
5.234	1	5.170	1	1.908	3	1.894	1
4.245	2	4.208	1	1.898	4	1.887	3
		4.015	1	1.804	3	1.794	2
3.557	3	3.527	1	1.788	2		
3.319	8	3.279	8	1.763	1		
		3.236	2	1.748	1	1.739	1
3.150	9	3.107	9	1.734	4	1.725	3
2.993	10	2.968	10	1.685	4	1.670	3
2.839	10	2.836	10	1.596	3	1.585	2
2.830	1	2.799	4	1.591	3	1.580	3
2.793	2	2.762	3			1.558	2
2.762	7	2.744	5			1.554	2
2.713	6	2.695	6			1.541	1
2.553	3	2.544	2	1.547	2	1.528	2
2.487	4	2.472	5	1.517	1	1.507	1
2.423	3	2.378	1	1.495	2	1.489	2
2.312	1			1.478	1	1.474	2
2.283	1			1.472	1	1.464	1
2.261	3	2.238	2	1.441	1		
2.212	5	2.197	5	1.437	1	1.426	1
2.124	1	2.111	2			1.417	2
2.093	4	2.071	3	1.395	1	1.385	3
2.073	4	2.057	3	1.386	1	1.376	1
1.994	2	1.979	3	1.288	3		
1.943	4	1.922	1				

with these. The zones have been formed by a later supply of alkalis, fluorine and water. Due to reasons outside the scope of this paper, it is most convenient to connect this supply of material with the formation of surrounding gneisses. Thus the formation of wagnerite, purely genetically, appears to be of a contact-pneumatolytical or hydrothermal character.

ACKNOWLEDGEMENTS

The author wishes to convey his thanks to Persbergs Grufve AB for defraying the cost of the analyses, to Prof. F. E. Wickman, Riksmuseet, for reading the manuscript, and to Mr. L. Jansson for carrying out the mineral separations.

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Tryckt den 10 september 1956

Uppsala 1956. Almqvist & Wiksells Boktryckeri AB

Silurian Echinoids from Gotland

By GERHARD REGNÉLL

With 4 plates and 4 figures in the text

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Abstract

Remains of echinoids are extremely rare in the fossil record of the Silurian of the Island of Gotland, Sweden. The material examined by the present writer includes a fragment of a test attributed to *Gotlandechinus balticus* n. gen., n. sp. (fam. Gotlandechinidae n. fam., order Melonechinoida MORTENSEN); some detached cidaroid radioles forming the basis for the description of *Silurocidaris clavata* n. gen., n. sp. (? fam. Archaeocidaridae M'COY); spiniform radioles not precisely determinable; and, finally, parts of Aristotle's lantern some of which are referred to *Lanternarius*

latens n.gen., n.sp. (inc. fam., inc. ord.). No pre-Carboniferous Melonechinoida have been known safely before, nor have any pre-Devonian Cidaroida. In connexion with the descriptions some remarks are made on the early phylogeny of the echinoids.

1. Introduction

Remains of echinoids are among the truly rare objects of the fossil record of the Paleozoic, and particularly so where the pre-Devonian, or, perhaps more correctly, the pre-Neodevonian section of the stratigraphic column is concerned.

Cambrian echinoids are unknown throughout; Ordovician and Silurian forms recognized up to 1937 were listed by MacBride & Spencer (1938, pp. 92–93). According to these authors, there are three Ordovician genera, viz. *Myriastiches*, *Aulechinus*, and *Ectinechinus*, each with one species—all from Britain—and, further, *Bothriocidaris*, from Esthonia. MacBride & Spencer were, however, inclined to adopt Mortensen's opinion of *Bothriocidaris* as no true echinoid but as a highly specialized derivative from some diploporite cystoid. At any rate, there is very strong evidence in favour of Mortensen's theory (cf. Mortensen 1935, p. 3 *et seq.*; also Termier & Termier 1953, p. 878), but it should not be overlooked that the *Bothriocidaris*-theory also has prominent advocates (Hawkins 1943, p. LXIII).

Five genera of Silurian Echinoidea were recorded by MacBride & Spencer (1938), including the following species: "*Wrightia*" *phillipsiae* (FORBES, 1848)¹ from near the Malvern Hills, Welsh Borderland, Upper Valentian; *Lepidocentrus*? sp.² (lantern only), from Dudley, Wenlock Limestone; *Echinocystites pomum* WYVILLE THOMSON, 1861, from Leintwardine, Herefordshire, Lower Ludlow; *Palaeodiscus ferox* SALTER 1857 (as the preceding species). Besides these British species there is also a North American one, namely *Koninckocidaris silurica* JACKSON, 1912,³ from Rochester, New York, base of the Rochester Shale, Clinton.⁴

For the sake of completeness it should be added, in agreement with Jackson (1912, p. 236, foot-note 2), that reference was made by Austin (1848, pp. 293–294) to "a large species of *Echinocrinus* [*Archaeocidaris*]... in the Wenlock limestone of Gliddon Hill" Shropshire. It was said to differ from congeneric Carboniferous forms in "having wider ambulacra, and a greater number of plates in the interambulacral spaces". However, the actual specimen was not described in any detail. If still preserved, its present depository is unknown.

¹ As observed by MacBride & Spencer (1938, p. 93), this species represented by a "well-nigh indeterminable" fragment (Hawkins & Hampton, 1927, p. 598), was referred by Jackson (1912, pp. 311, 312, 316) to *Maccoya*, and by Hawkins & Hampton (1927, pp. 597–598) tentatively to *Echinocystites*. Mortensen (1935, p. 38) treated *Wrightia* as a synonym of *Maccoya*. Under the name of *Wrightella* POMEL, 1883, *Wrightia* was made a sub-genus of *Maccoya* by Lambert & Thiéry (1910, p. 120). The genus *Wrightia* POMEL, 1869, is obscure also from a nomenclatorial point of view, being a homonym of *Wrightia* AGASSIZ, 1862 (Coelenterata). On account of this, *Wrightia* had to be rejected and was replaced by *Wrightella* POMEL, 1883. However, this name, too, is invalid, being pre-occupied by *Wrightella* GRAY, 1870 (Coelenterata) (see Neave 1940, p. 659).

² As appears from Jackson's (1912, p. 236, foot-note 1) comment on the single specimen known, the generic attribution (made by Jackson) is loosely founded.

³ According to Mortensen (1935, p. 58), this species is not likely to belong to *Koninckocidaris* but rather to *Aulechinus* (Ashgill) or *Palaeodiscus*. Even the status of the genus *Koninckocidaris* DOLLO & BUISSET, 1888, is said to be doubtful.

⁴ The Rochester Shale can be correlated broadly with strata on the Valentian (or Llandovery) –Wenlock boundary in Great Britain (Swartz *et al.*, 1942, chart).

The author is not aware of any other addition to be made to the above list of Silurian echinoids, apart from the material from Sweden to be dealt with below.

The Lower Devonian has so far yielded only a single determinable specimen which was described by Dehm (1953) as *Rhenechinus hopstättereri*. This unique find comes from the Hunsrück Shale at Gemünden, Germany. In addition, this formation has yielded isolated interambulacral plates designated as *Tornquistellus* (about which see Mortensen 1935, p. 65).

Echinoids are very scarce also in Middle Devonian rocks. A species of *Lepidechinois* was recorded by Cooper (1931, p. 132) from the Hamilton group (of Givetian age) in the State of New York; isolated spines of *Xenocidaris* SCHULTZE, and a couple of species of *Lepidocentrus* J. MÜLLER are known from the Rhineland, Germany (Jackson 1912, pp. 454, 286). About *Palaeochinus* recorded from Morocco, see p. 176 below.

2. The fossil material from Gotland

It is evident from the above review that the famous fossil record of the Silurian of Gotland, Sweden, has not contributed any form to the select company of Old Paleozoic echinoids described from various parts of W. Europe and N. America. Yet the occurrence of echinoids in the Silurian of Gotland cannot be said to be entirely new to science. References to such fossils have been made in the literature but have been largely overlooked, presumable owing to the fact that no information on the actual nature of the fossil material has been available.

In the "List of the fossils of the Upper Silurian formation of Gotland" edited by G. Lindström (1885) the following entry is found on p. 14 under the heading of Echinoidea: "Parts of the dental apparatus of an Echinoidean have been found in several places." A few years later (Lindström 1888, p. 16), some additional information was given: "Parts of the dentary apparatus of an Echinoidean and probably also plates and spines of two other different species have been found in the strata f-h."¹

Hedström (1910, pp. 1467, 1472) recorded "Echinoidean thorns" (*sic*) from a section known as the "Vattenfallet" ("Cascade") section in the so-called Palisades south of the town wall of Visby. The echinoid spines were obtained partly from what is now called the Upper Visby marl, and partly from beds belonging to the Höglint group.

This seems to be all that has been published about the Silurian echinoids of Gotland. As we have seen, the oldest of these printed sources dates from 1885. But it is interesting to note that "Echinittaggar" (echinoid spines) from Gotland are mentioned in three instances in a catalogue of the mineral collections of the Council of Mines (Bergskollegium) written by hand by P. S. Hjelm and completed in 1803.² The specimens referred to were probably amongst fossils deposited in the Paleozool. Dept. of the Swedish Museum of Natural History in 1867. But, if still preserved, they can no longer be identified.³

¹ These strata form the so-called "Upper Limestone group" of Lindström; cf. Hede, 1921, p. 12 *et seq.*

² This catalogue is now in the archives of the Mineralogical Dept. of the Swedish Museum of Natural History, Stockholm. Dr. N. Zenzén (*in litt.*) very kindly directed the writer's attention to the entries on the Gotland echinoid spines.

³ In the older literature there are numerous references to "echinoids", by which are meant cystoids, particularly *Echinospaerites* and *Sphaeronites*. A special case is represented by the Lower Cambrian *Spatangopsis costata* TORELL (1870, p. 11) which was interpreted tentatively as an echinoderm and, as is evident from the generic name, as an echinoid. This fossil is now known as *Medusina costata*.

All material at the writer's disposal belongs to the above-mentioned Institute (for which the abbreviation RM will be used below). It comprises three groups of echinoid remains, namely plates, isolated spines, and elements constituting Aristotle's lantern. Unfortunately, these various parts are generally dissociated so that their relations to each other are obscure. It is also to be regretted that there is only one minor fragment of the test, since the latter, on the whole, possesses more significant features than the spines and jaw elements.

3. Description and discussion of fossils

Students of Paleozoic echinoids are fortunate in having at their disposal two indispensable monographs which provide a firm basis of all subsequent work in this field, viz. Jackson's magnificent "Phylogeny of the Echini" (1912) and Mortensen's monumental "Monograph of the Echinoidea" (1928, onwards); the latter treats living as well as fossil forms. The morphology of the Paleozoic genera is more fully dealt with by Jackson, for obvious reasons, but the classification advocated by him has been improved in important respects by Mortensen. The classification proposed by Mortensen will be adopted in the present paper.

A. Order MELONECHINOIDA MORTENSEN, 1934

In the genus to be described below the coronal plates are not imbricating, they are disposed in more than 20 columns, and the tubercles are small. Hence it must be referred to the Melonechinoida which have hitherto comprised Lower Carboniferous forms only. As conceived by Mortensen, the order contains one family only, Palae-echinidae M'COY, 1849, the characters of which are the same as those of the order.

In his key to the genera of the Palae-echinidae, Mortensen (1935, p. 36) recognized two main groups, one having "ambulacra with only two columns of plates, all reaching from the median line to the edge of the area",¹ the other one having "ambulacra with four or more series of plates, none of them (at the ambitus) reaching from the median line to the edge of the area". The specimen under consideration which represents a new genus does not fit into any of these categories. Accordingly, a new family has to be created to receive the new genus, whereas the characters just quoted should apply to the family Palae-echinidae.

Family *Gotlandechinidae* n. fam.

FAMILY TYPE.—*Gotlandechinus* n. gen.

DIAGNOSIS.—A family of Melonechinoida with two columns of plates in each ambulacral area, every second plate in each half-ambulacrum reaching from the perradial suture (median line) to the adradial suture (edge of the area), alternating with occluded plates; pore-pairs uniserial.

Genus *Gotlandechinus* n. gen.

GENOTYPE.—*Gotlandechinus balticus* n. sp.

DERIVATION OF NAME.—From Gotland, in allusion to the region yielding the actual specimen and Lat. *echinus*, sea urchin.

¹ This is not unconditionally true of one genus of the Palae-echinidae, viz. *Maccoya* POMEL, in which certain plates of the ambulacral mid-zone may be completely occluded, as seen from the exterior (in an interior view all plates cross the half-areas; Jackson 1912, p. 311).

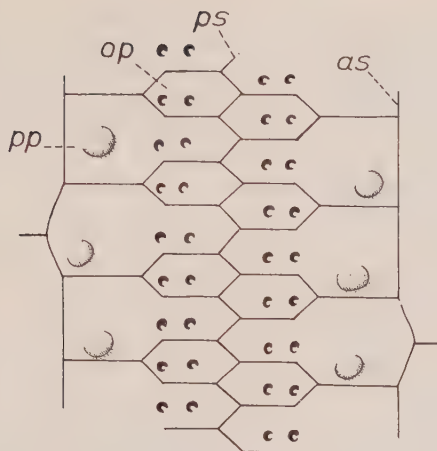


Fig. 1. *Gotlandechinus balticus* n.gen. n.sp., portion of ambulacrum, slightly diagrammatic. *as*, adradial suture; *op*, occluded plate; *pp*, primary plate; *ps*, perradial suture. $\times 7.5$ (approx.).—Sweden, Gotland.

DIAGNOSIS.—A genus of Gotlandechinidae with very regularly disposed ambulacral plates. (There are four columns of plates in each interambulacrum, but this character is deemed to be on the specific level rather than on the generic one.)

DISCUSSION.—*Gotlandechinus* being for the present the sole representative of Gotlandechinidae, the distinctive features of the family are valid for the genus as well. Thus it differs from all genera of the Palaeochinidae in the shape and arrangement of its ambulacral plates (text-fig. 1). A cursory survey will show the main differences and similarities between *Gotlandechinus* and other genera of the Melonechinoida. As mentioned already, all of these flourished in early Carboniferous time.

Palaeochinus (W. Europe, N. Africa? and N. America¹) is, in fact, a more primitive type with regard to the ambulacral structure than the geologically much older *Gotlandechinus* in that the plates of each half-ambulacrum are uniserial. Also the pore-pairs are uniserial in both genera, due to a very special condition of the pores of the adradial plates in *Gotlandechinus* (see below under the description of the holotype). As in the Swedish genus, four columns of interambulacral plates are found in a couple of European species of *Palaeochinus*, while in others there are five (in *P. canadensis* eight, Kier 1953, p. 67).

In *Maccoya* also (W. Europe, N. Africa,² and N. America) there are a few species which are characterized by four columns of interambulacral plates. Typically, all plates of an half-ambulacrum extend from the perradial to the adradial suture, but cf. above, p. 158, foot-note 1. The pore-pairs are biserial.

In *Lovenechinus* (W. Europe and N. America), *Oligoporus* (N. America), and *Melonechinus* (USSR, W. Europe, and N. America) there is a progressive differentiation towards greater complexity of the ambulacra, as compared with *Palaeochinus* and *Maccoya*. This fact was displayed by Jackson (1912, p. 54) and Mortensen (1935, p. 30). The number of columns of interambulacral plates varies in the three genera mentioned,

¹ *P. canadensis* KIER, 1953, is the first certain member of the genus to be known from N. America. Echinoid remains from the Visean of Morocco were assigned to *P.* by Termier & Termier (1950, p. 98).

² *M.* sp. from the Visean of Morocco (Termier & Termier 1950, p. 98).

but all of them include species which have four columns of interambulacral plates (in *Oligoporus* this number seems to have been noticed only as a teratological phenomenon; cf. Jackson 1912, p. 352).

In various species belonging to the Palaeoechinidae there is a tendency of the test to be elevated into meridian ribs (Mortensen 1935, p. 29). This tendency is not apparent in *Gotlandechinus*.

REGIONAL DISTRIBUTION.—Sweden (Gotland).

STRATIGRAPHIC RANGE.—Upper Silurian.

Gotlandechinus balticus n. gen. n. sp.

Pl. 1, fig. 4. Text-figs. 1-2.

DERIVATION OF NAME.—*balticus*, from Medieval Lat. *Mare Balticum*, the Baltic.

HOLOTYPE.—RM Ec. 68.

TYPE LOCALITY.—Klinteberget, E. of the church of Klinte, Gotland.

TYPE STRATUM.—Yellowish grey crinoidal limestone of the Klinteberg group, Upper Silurian, probably Lower Ludlow.¹

MATERIAL.—The holotype which is the only specimen available is a very incomplete corona, a portion of an interambulacrum, part of two plates of another interambulacrum, and portions of two adjacent ambulacra being preserved. The fragment which is seen in an outer (distal) face measures approximately 28 by 28 mm. It is slightly vaulted, extending probably from the region of the ambitus towards the placogeneuous zone.² This interpretation is based on the fact that, when the fragment is orientated accordingly, the outer pore of a pore-pair is situated slightly higher than the inner pore as is usual in Paleozoic echinoids. The position of the fragment in relation to the antero-posterior axis cannot be determined.

DIAGNOSIS.—A species of *Gotlandechinus* with four columns of plates in each interambulacrum; on an average, three ambulacral plates border upon one interambulacral plate on the adradial suture. (The fossil material available does not permit a full diagnosis.)

DESCRIPTION.—The fragment gives no idea of the *general shape* of the corona, but its vertical diameter may be estimated at 40-50 mm.

One of the *ambulacra* is preserved to a length of 24 mm. Almost throughout its whole extent it has a practically uniform width of 6 mm, yet it seems to taper slightly in an adapical direction. This means that in the midzone the width of the ambulacra is about one third of that of the interambulacra. The whole structure is not elevated above the level of the adjacent interambulacrum.

The *ambulacral plates* are dimorphic and very uniformly developed along the whole length of the ambulacrum. All plates are simple, as in other Paleozoic echinoids, and meet in a regular perradial zigzag line. In each half-ambulacrum there is a regular alternation between so-called primary plates reaching from the perradial suture to the adradial suture and so-called occluded plates reaching from the perradial suture about

¹ The Klinteberg group is underlain by the Mulde marl which according to Hede (1942, p. 226 [p. 22 of sep. print]) probably has to be co-ordinated with the zone of *Cyrtograptus lundgreni* in the British standard section. The Klinteberg group is overlain by the Hemse marl which "is to be correlated with the *Monograptus nilssoni* zone of the Lower Ludlow (Hede, l.c.). Přibyl (1943, p. 1) advocated a Lower Ludlovian age of the Klinteberg group.

² "The placogeneuous zone ... is that portion of the corona next to the apical [aboral] disc in which the newly added coronal plates occur both in the ambulacrum and the interambulacrum" (Jackson 1912, p. 26).

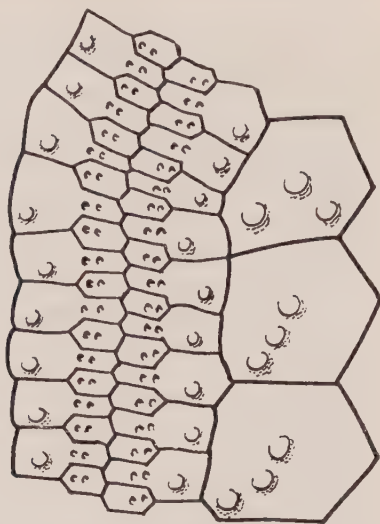


Fig. 2. *Gotlandechinus balticus* n. gen. n. sp., portion of ambulacrum, with adjacent adradial column of inter-radial plates. $\times 5$. — Sweden, Gotland.

midway to the adradial one (see text-fig. 2).¹ The primary plates can be said to be more or less bottle-shaped, with an approximately flat "bottom" at the adradial suture. The "neck" of the "bottle" is of about the same length as the "trunk" and of about half the width of the latter. In general, three primary plates — or rather two plates and the adjacent halves of two more primaries — equal the height of an interambulacral plate in the adradial column, but variations occur. The occluded plates, located in between the "necks" of the primary plates, are very regularly hexagonal. Their width is about 1.5 mm, whilst their height is about half their width.

The *pore-pairs* are uniserial, which means that the pores of the primaries are located in the "neck" of these bottle-shaped plates. The outermost pore of a pair is very nearly on a line with the inner pore, or very slightly higher. Peripores can scarcely be recognized but may well have existed and become secondarily obliterated owing to slight wearing of the test. According to Jackson (1912, p. 57), the pore-pairs in all Paleozoic genera, apparently without exception, are enclosed in a peripore.

A small imperforate (secondary) *tubercle* seems to be present in each primary plate of the ambulacrum, being located in the widened portion of the plate.²

The *interambulacrum* is made up of four regular columns of *interambulacral plates*. These are moderately thick — approximately 0.8 mm — and border edgewise upon each other. Those in the mid-columns are hexagonal. In the mid-zone the plates are slightly more wide than high, while the relative height seems to increase in an adapical direction, at the same time as the absolute size of the plates decreases in connexion with the gradual tapering of the interambulacral area. The plates in the adradial columns are sub-pentagonal but otherwise of the same general type as the plates mentioned. For example, the largest plate present is about 5 mm wide and 4 mm high.

¹ On a superficial examination, this feature of the plates may simulate a quadriserial arrangement of the ambulacral plates. Notwithstanding the differentiation of the ambulacral plates into two morphological types, there can hardly be any objection to interpreting each half-ambulacrum as uniserial, the more so as the pore-pairs are uniserial.

² In fig. 4 on Pl. 1 the tubercles do not show up clearly either in the ambulacral or in the interambulacral plates. They are best visible on a strongly oblique illumination.

Tubercles for (secondary) spines similar to those in the ambulacral plates are found in the interambulacral plates as well; they seem to vary from three to five in each (cf. text-fig. 2).¹

DISCUSSION.—In recognition of the monotypy of *Gotlandechinus*, a taxonomic comparison with other species is not required, but reference may be made to the brief discussion above of other genera of the Palaeochinidae.² On the other hand, the uniqueness of the find invites some consideration from a morphological point of view.

Obviously, the greatest interest is attached to the differentiation of the ambulacral plates into primaries and ocluded plates. It is reasonable to assume — as do all students of Paleozoic echinoids — that the primitive arrangement of the ambulacral plates is that found in *Palaeochinus*, namely one column of uniform plates in each half-ambulacrum, as in the Late Ordovician *Lepidocentridae*. As indicated above, it may seem remarkable that a more advanced stage should be represented by the Silurian *Gotlandechinus*. However, an analysis of the stratigraphic appearance of forms having only primary ambulacral plates and forms having primaries alternating with ocluded plates reveals that the latter type is also of ancient origin. Probably it branched off in early Silurian or even perhaps, in Ordovician times. Subsequently, the two kinds of ambulacral structure persisted side by side both in the Palaeochinidae and the *Lepidocentridae*. That this is so will appear from Table 1 below, in spite of the fact that only fragments of the different lines of development are documented in the fossil record. Genera possessing demi-plates as well are not included in the table.

Uniseriality of ambulacral pore-pairs is characteristic of ambulacra made up of simple primaries. But uniseriality of pore-pairs is exceptional in genera with prima-

Table 1.

	Only primaries	Primaries alternating with ocluded plates	Only primaries	Primaries alternating with ocluded plates
L. Carb.	<i>Maccoya</i> , <i>Palaeochinus</i>	<i>Maccoya</i> (mid-zone)	<i>Lepidocentrus</i> a. o.	<i>Pholidechinus</i> (midzone)
U. Dev.			<i>Lepidechinoides</i>	
M. Dev.			<i>Lepidechinoides</i>	
L. Dev.				<i>Rhenechinus</i>
U. Sil.		<i>Gotlandechinus</i>	<i>Palaeodiscus</i>	<i>Echinocystites</i>
M. Sil.		<i>Maccoya philipsiae</i> (cf. p. 156)		
U. Ord.			<i>Aulechinus</i> a. o.	
	Palaeochinidae		Lepidocentridae	

¹ The dark canals visible *inter alia* in the lower plates of the left interambulacral column are probably due to injury inflicted upon the plates by the action of some boring organism.

² It may be mentioned that the sample bearing the holotype (RM Ec. 68) is accompanied by a label in the handwriting of Th. Mortensen. The text is as follows: "*Palaeochinus quadriserialis* WRIGHT? (n. sp.?)". It is, of course, very unlikely *a priori* that an echinoid from Gotland would be conspecific with a specimen from the Lower Carboniferous. On examination of the Gotland specimen it is not only useful but necessary to use refractive fluids to get a correct idea of the structure of the ambulacrum. Mortensen evidently did not do so, or he would not have suggested an identification with *Palaeochinus* in which ocluded ambulacral plates do not occur.

ries alternating with occluded plates. It is significant that in *Pholidechinus* the pore-pairs are slightly biserial at the mid-zone, where there is a tendency for primary plates to alternate with occluded ones, whilst otherwise the pore-pairs are uniserial (Jackson 1912, p. 299). The uniseriality of the pore-pairs in *Gotlandechinus* is due to the fact that the pores of the primaries approach closely to the perradial suture so as to be brought in line with the pore-pairs of the occluded plates, the entire width of which is only about half of that of the primaries. According to Jackson (1912, p. 57), the pores do not "lie nearer the median suture of the ambulacrum than the middle of the ambulacral plate" in any Paleozoic form. However, in *Cravenechinus* HAWKINS, 1946, from the Viséan of Yorkshire, the pore-pairs are perradiad in the adradial column of ambulacral plates (the ambulacra are multiserial), which "carry their pores at the 'wrong end' when judged by normal Echinoid standards" (Hawkins 1946, p. 194). "The wide separation of the outer pore-rows from the adradial suture is apparently unique. The only case of a perradiad position of the pores in the adradial column known to me is that figured by Jackson (1912, pl. LXVII, fig. 5) in *Proterocidaris*" (Hawkins 1946, p. 196). But Hawkins added that "since the figure shows the inner surface, the position of the pores does not necessarily correspond with their external situation". A critical appraisal of what is actually known about *Proterocidaris* prompts one to caution in comparing the position of its ambulacral pores with that of *Gotlandechinus* and *Cravenechinus*.¹ In the latter genus, there is a single row of interambulacral plates. Hawkins (1946, p. 196) suggested that the strength of the test was increased by incorporation of the imperforate adradial parts of the ambulacra with the very narrow interambulacral regions. This idea, plausible in the case of *Cravenechinus*, is hardly applicable to *Gotlandechinus* in which the interambulacral fields are broad, and the significance of this feature remains obscure.

The nature of the interambulacral areas in *Gotlandechinus* hardly requires any comment. Mortensen (1935, p. 31) pointed out that there are four columns of interambulacral plates in the more primitive species within all the genera of the Palaeoecchinidae, except *Melonechinus*.

Summing up, *Gotlandechinus balticus*, as might be expected, is characterized by a number of primitive features, including biseriality of ambulacral pore-pairs, and quadriserality of interambulacral plates.²

REGIONAL DISTRIBUTION.—Sweden, Gotland: Klinteberget.

STRATIGRAPHIC RANGE.—Upper Silurian, probably Lower Ludlow.

¹ Jackson's (1912) description and figures of *Proterocidaris* are based entirely on information derived from the literature, particularly from Fraipont (1904). On what observation Jackson (1912, p. 411) founded his statement that the ambulacral "pore-pairs lie near the middle of each half-area, doubtless due to the fact that it is an internal view" the present writer does not know. In describing *Oligoporus soreili* FRAIPONT, 1904, which, as shown by Jackson, is identical with *Proterocidaris giganteus* KONINCK, 1881, Fraipont (1904, p. 10) remarked that he had not succeeded in making out the exact ambulacral structure, and his Pl. 5, fig. 2, on which is based Jackson's Pl. 67, fig. 5, referred to by Hawkins, gives no idea of the position of the pore-pairs, neither do other figures communicated by Fraipont. Later on, Jackson had an opportunity to study extensive material of *Proterocidaris*. In his monograph of the "Palaeozoic Echini of Belgium" Jackson (1929, p. 54) stated categorically that the "pores of ambulacral plates are nearer the interambulacral suture than they are to the middle of the plate".

² In *Tetracidaris* COTTEAU this "Palaeoecchinoid character [four columns of plates in each interambulacrum] is associated with a type of ambulacrum which, for the Cidaridae, is remarkably specialised" (Gregory 1900, p. 303). Mortensen (1928, pp. 52, 527) favoured the opinion that this Eoeretaceous genus is a highly specialized offshoot from *Diplocidaris*, having no close connexion with the Archaeocidaridae.

B. Detached radioles

Unfortunately, no spines have been found in association with *Gotlandechinus*, nor, for that matter, in beds at any other locality equivalent to those bearing *Gotlandechinus* at Klinteberget. But a number of isolated spines give evidence of the presence of echinoids in different horizons of the Silurian sequence on Gotland.

It is an established fact that isolated echinoid spines are stubborn objects from a taxonomic point of view. The difficulties increase with increasing antiquity of the forms to be examined. Even when dealing with a material as relatively "recent" as Triassic radioles, Bather (1909, p. 59) had to admit that "so far as these Triassic remains are concerned, it is rarely, if ever, possible to be sure of the genus to which a radiole or a jaw-element belongs". Jackson (1912, p. 78) stated that there are only small-sized, secondary spines in the Palaeochinidae, and further, that as "far as known, spines are very uniform in character within the species in the Palaeozoic". He therefore considered the spines to provide "very good species characters". This may be true, but the statement has certainly no universal applicability.

Histological features — so useful in the characterization of Mesozoic and younger radioles — are of little service in dealing with Paleozoic, and particularly Old Paleozoic, forms. Hesse (1900) who made the only existing special investigation into this matter, examined one Devonian and two Carboniferous species, but did not have access to any further Paleozoic material. Termier & Termier (1950, Pl. 232, fig. 5) communicated a figure of a thin section of a Devonian radiole attributed to *Palaeochinus*, and some other similar figures may have been published. In many cases, however, the detailed structure of the Old Paleozoic radioles has undoubtedly been obliterated, more or less completely, by processes of recrystallization.

It is obvious that spines were feebly developed — setaceous or nearly so — in most Early Paleozoic echinoids, although the tiny spines could be very numerous, as in the Silurian *Palaeodiscus* (see also Hawkins 1943, p. LXV). According to Mortensen (1935, p. 56), spines are apparently entirely wanting in the Late Ordovician *Myriastiches*. If this holds good, *Myriastiches* is quite extraordinary in this respect, because spines are characteristic of echinoids and "there is no known species which is without them" (Jackson 1912, p. 77).

In the material available from Gotland there are two main types of echinoid radioles, namely claviform ones, deemed to be cidaroid, and spiniform ones.

Cidaroid radioles

The present writer has been very much in doubt whether a generic and specific name should be applied to these detached spines, or not. The decision to throw his scruples to the winds was based on the following considerations: although incomplete, the spines are clearly defined morphologically; they are represented by several specimens; there are no coronal remains with which the spines could possibly be combined; if not fixed by a name by the present writer, the spines are very likely to be named by a subsequent reviser, who may not have access to the actual fossil material.¹

¹ Bather (1909, p. 7) was faced to a similar problem with respect to isolated columnals of crinoids. His comment was: "As regards the application of specific names to such fragments, it seems to me that if our descriptions are to be of practical service to stratigraphers, then they must be accompanied by names. A description without a name is soon lost sight of, whereas a name compels attention until at last it finds its proper position, if only as a synonym." In this connexion, see also Bather (1909, pp. 244–245).

? Family *Archaeocidaridae* M'CoyGenus *Silurocidaris* n. gen.

GENOTYPE.—*Silurocidaris clavata* n.sp.

DERIVATION OF NAME.—*Silurocidaris*, the Silurian *Cidaris*, this genus being the only (presumably) cidaroid form hitherto recorded from Silurian beds.

DIAGNOSIS (with reference to radioles only). — A genus of *Archaeocidaridae* (?) having primary spines with a globular, or subconical, shaft, finely and densely striated longitudinally, and with numerous minute verruciform elevations all over the surface.

DISCUSSION.—As mentioned already, these radioles are so far the only ones of this kind known from Silurian beds. There is not much doubt that they belong to the *Cidaroida*, but, on the other hand, this can hardly be demonstrated conclusively. It is true that, in their general type they conform with cidaroid spines, but thin sections (Pl. 4, figs. 3–5) do not show the microstructure characteristic of the *Cidaroida*, that is to say a cortex, and a layer of radiating septa surrounding a core formed by an irregular calcareous meshwork (see e.g. Mortensen 1928, p. 24). A differentiation into three layers is not apparent in the slides. Rather it seems that the radiole is made up of a uniform tissue of regularly disposed longitudinal elements and more irregular radiating ones. As all specimens available, save the holotype of *Silurocidaris clavata* n.sp., are considerably abraded, it is quite conceivable that a cortical layer was present originally. Some observations by Hesse (1900, p. 212) on thin sections of the Middle Devonian *Xenocidaris clavigera* (see below about the relations between *Silurocidaris* n.gen. and *Xenocidaris*) may have a certain interest in this connexion. The state of preservation of the radioles was said to be rather poor. A cortex was not preserved, but the cidaroid nature of the spines was evident from the presence of an "axial complex" of fairly thick-walled tubes surrounded by a zone of radiating septa. In the case of *Silurocidaris*, the microstructure is much less significant, and for that reason this genus is not unreservedly attributed to the *Archaeocidaridae* (see also p. 167 below, foot-note 1).

This attribution, however, finds a certain support in the resemblance which can be claimed to exist between *Silurocidaris* and *Nortonechinus* THOMAS. Four species of this genus were described by Thomas (1920) from the Upper Devonian¹ of Iowa. In both genera, the radioles are greatly expanded distally, a feature which, as pointed out by Thomas (1920, pp. 482–483), is rare in early echinoids. "In the Paleozoic such development was heretofore unknown barring the moderate distal expansions of *Xenocidaris*."²

¹ The Lime Creek beds which have yielded an abundant material of dissociated plates and spines, a part of a broken test, and parts of the lantern of *Nortonechinus* (cf. Thomas 1920, p. 481) form an upper part of the Senecan (Schuchert 1943, p. 700) which is generally supposed to be an equivalent of the Frasnian in Europe.

² Mortensen (1928, p. 60) suggested that *Nortonechinus* THOMAS, 1920, be a synonym of *Xenocidaris* SCHULTZE, 1866. The latter is represented by isolated spines from Middle Devonian beds in the Rhineland, Germany, and by various skeletal remains from the Upper Devonian of Belgium (Maillieux 1940, p. 32) and Iowa (Thomas 1920, p. 496). The spines are of the same general type, and the difference in shape was not considered by Mortensen important enough to serve as a generic distinction. Thomas (1920, p. 482), on the other hand, maintained that the resemblance is casual and "more likely a case of parallel adaptation than of close relationship". Maillieux (1940, pp. 31–32) discussed the matter of the relationship between *Xenocidaris* and *Nortonechinus*. He recognized the close affinity between these forms but thought it wiser to retain both genera provisionally until more complete material of *Xenocidaris* would make possible a definite decision. It may be added that reference was made to *Xenocidaris* (with some hesitation assigned to the

In the radioles of *Nortonechinus* the apical surface is flattened, pustulose, or slightly spinulose. Also certain specimens of *Silurocidaris* are distinctly flattened distally, for example those figured in Pl. 1, fig. 3, and Pl. 2, fig. 1. Apical spinules, which are still more strongly developed in *Xenocidaris*, were certainly not present in *Silurocidaris*. In transverse section the radioles of this genus are circular, or subcircular. *Nortonechinus* shows a higher degree of specialization in that the apical surface is very often polygonal due to mutual contact caused by crowding of the spines. This implies that, in *Nortonechinus*, the spines probably covered the corona completely, as in *Anaulocidaris* (Cidaridae, Streptocidarinae; Triassic), *Acrocidaris* (Pseudodiademmatidae; Jurassic-Cretaceous), and *Colobocentrotus* (Echinidae; Recent) (cf. Bather 1909, pp. 94, 169-170; Thomas 1920, p. 491 *et seq.*; Mortensen 1928, pp. 37-38, 60; Cuénot 1948, pp. 178, 185). *Colobocentrotus* lives on rocks exposed to the battering of the surf. This will also have been true of those older forms provided with a protective cover of a similar kind. Whether *Silurocidaris* was a reef-dweller or not is hardly apparent from the morphology of the radioles, but at both localities from which specimens have been obtained reef-limestone is exposed, according to the geological map-sheets.

Among Mesozoic forms having radioles reminiscent, more or less, of those in *Silurocidaris* may be mentioned *Balanocidaris? dorsata* (BRAUN) (Triassic) (Desor 1858, Pl. 2, fig. 4), several Jurassic species of *Balanocidaris*, and *B. pleracantha* (AGASSIZ) (Cretaceous) (Desor 1858, Pl. 6, figs. 7-9.)

REGIONAL DISTRIBUTION.—Sweden (Gotland).

STRATIGRAPHIC RANGE.—Upper Silurian.

Silurocidaris clavata n. gen. n. sp.

Pl. 1, figs. 1-3. Pl. 2, figs. 1-2. Pl. 4, figs. 3-5.

DERIVATION OF NAME.—From Lat. *clavatus*, club-shaped.

HOLOTYPE.—RM Ec. 71.

TYPE LOCALITY.—Manngårde, Lye, Gotland.

TYPE STRATUM.—Limestone¹ of the Hemse group, Upper Silurian, Lower Ludlow.²

MATERIAL.—Besides the holotype which is the only specimen available from the type locality, there are ten specimens in the Paleozoological Dept. of the Swedish Museum of Natural History. Three of these were sectioned.

DIAGNOSIS.—Same as that of the genus, by monotypy.

DESCRIPTION.—As frequently with fossil radioles, the shaft³ is detached along the syzygal suture against the collarette, so that the proximal portion of the spine is unfortunately not available for study. From the fractural surface which is circular or subcircular in transverse section, the blade expands so as to form an inversely onion-like body. In an apical view, this body may be somewhat assymetrical, but is largely circular, or subcircular. The greatest diameter of some specimens appears from the following table (measurements in mm).

Cidaridae) in Zittel-Broili (1924, p. 262), while *Nortonechinus* was not mentioned. The reverse applies to Termier & Termier (1953, p. 888) who placed *Nortonechinus* with the Archaeocidaridae.

It may be mentioned parenthetically that spines with an apical portion closely resembling that in *Xenocidaris* is found in *Cyathocidaris? avenionensis* DESMOULINS (see Desor 1858, Pl. 7, fig. 8), and in other species of this Tertiary genus (Mortensen 1928, p. 484, fig. 150).

¹ On the geological map-sheet "Ronehamn" the exposure just N. of Manngårde is laid out as reef-limestone.

² According to Hede (1942, p. 226 [22 of sep. print]), the Hemse marl belongs to the zone of *Monograptus nilssoni*, of the Lower Ludlow.

³ The terminology employed in describing the radioles is adopted from Bather (1909, p. 135).

RM Ec. 70a1	RM Ec. 70c1	RM Ec. 70c3	RM Ec. 71 (holotype)	RM Ec. 70a2	RM Ec. 70c2	RM Ec. 70b
7.8	7.8	6.6	6.2	6.0	6.0	4.4

There is a certain variation in the general outline of the radioles in a lateral view (cf. the figures). In some specimens, the distal expansion is more spherical; in others, it is more conical due to a distinct flattening of the apical surface. In all instances, the margin between the sides and the apical surface is smoothly rounded and shows no indications of having been faceted.

In the holotype (Pl. 2, fig. 2), the surface is raised all over into minute, flattened, verruciform elevations. The whole specimen is ornamented with extremely faint longitudinal ridges (10–12 in 0.5 mm). The striation can be traced in RM Ec. 70a1 and 70a2 as well.¹ Moreover, a similar striation is also met with in *Nortonechinus* (Thomas 1920, p. 488, explanation of text-fig. 77; p. 494), and *Xenocidaris* (Maillieux 1940, p. 31).

DISCUSSION.—As should be evident from the above description, and from the figures, the radioles attributed to *Silurocidaris clavata* n.sp. vary to a certain degree. In view of the fact that all, apart from the holotype, originate from one locality and very likely from the same bed, the writer has found no reason to split them up into different species, the less so as the spines of later Cidaroids are known to vary within rather wide limits.

A further discussion of the relations of *Silurocidaris clavata* n.sp. to other species is hardly necessary, since the affinities and differences between the Gotland genus and *Nortonechinus* were dealt with at some length in a foregoing paragraph.

REGIONAL DISTRIBUTION.—Sweden, Gotland: Manngårde, Lye (the holotype); Lindklint (Linde klint) (paratypoids and autohyle).

STRATIGRAPHIC RANGE.—Upper Silurian, Lower Ludlow.

Spiniform radioles

The spiniform radioles available from the Silurian of Gotland are less indicative of their systematic status than the claviform ones, since they represent a much more commonplace kind of spines. The material is interesting in so far as it affirms the presence of echinoids in the beds from which it was derived, and adds to our scanty knowledge of echinoids as a noteworthy, though subordinate, element in the faunas inhab-

¹ The following observation by Mortensen (1928, pp. 26–27) has a certain bearing on our subject: "In the description of recent Cidarids it is very often mentioned that some of the Primary spines on the aboral side are very different in structure from the ambital spines, without spinules and very finely longitudinally striated. These are, however, not a different sort of spines, but only developmental stages of the usual primary spines. The longitudinal striation is due to the fact that the cortex-layer has not yet been formed, the radiating spokes thus reaching the surface; on the living specimens such spines will be found to be covered by a ciliated skin. When the spoke-layer has reached the definitive thickness, the cortex (or ostracum) begins to develop."

Applied to the Gotland material this means (1) that some, or all, of the spines may be immature, and (2) that, if this is so, we cannot expect to recognize the microscopical structure typical of cidaroids. In consequence, histological features would have no relevance at all in this case in judging the systematic position of the form carrying the spines. From a morphological point of view these are cidaroid. If little or no importance is attached to the microstructure there is no reason to doubt that the animal belongs to the Archaeocidaridae.

iting the seas covering Gotland in Silurian times. From a taxonomic point of view, however, this material has little interest. It would be useless to try to allocate the specimens to one family or other. Material from different stratigraphic units will be dealt with separately below.¹

a. Specimens from the Upper Visby marl

Pl. 3, fig. 3. Pl. 4, fig. 2.

At least three tiny spines are present in a small sample (about 15 by 18 mm; labelled RM Ec. 83b) from "Vattenfallet", S. of Visby. From the same locality there are also four isolated spines (RM Ec. 83a1-4) of similar appearance. They vary in length from about 4 to 7 mm, and are very slenderly fusiform, the greatest diameter being 0.55 mm in the figured specimen. There is no ornament.

In a thin section (prepared by G. Lindström?) of a small spine from "Wisby b", i.e. the Upper Visby marl, a faintly indicated lamellar structure can be traced at low magnification (Pl. 4, fig. 2). It recalls somewhat a thin section of a spine of *Porocidaris* (Recent) described by Mortensen (1928, p. 106).

As mentioned above (p. 157) echinoid spines were recorded by Hedström (1910, pp. 1467, 1472) both from the Upper Visby marl and from beds in the Höglint group at the locality called "Vattenfallet". Judging from the lithological character of the sample referred to above, it comes from the Upper Visby marl. The material is not identical with that listed by Hedström but belongs to older collections of the Paleozoological Department of the Swedish Museum of Natural History. It is labelled "Echinid taggar" (echinoid spines) in old handwriting (G. Holm?).

Spines of this kind occur in quite a number of Paleozoic echinoids.

The age of the Upper Visby marl is probably Upper Valentian (Hede 1942, p. 226 [22 of sep. print]).

b. Specimens from the Slite group

Pl. 2, fig. 3. Pl. 3, figs. 1-2. Pl. 4, fig. 1.

This material includes a number of samples (RM Ec. 73-77, some of them split up into several specimens marked with index letters) from Samsugn, 3 km S. of the church of Othem. A few other samples (RM Ec. 87a-h) lack data of horizon and locality, but agree perfectly with RM Ec. 75. They originate no doubt from the same locality. In RM Ec. 74 there is a fragment of a single spine only (associated with parts of a lantern), whilst in the other samples there are clusters of acicular spines. These are considerably longer than those available from the Upper Visby marl, the longest fragment reaching a length of 13 mm; a complete spine was undoubtedly much longer. Spines of the size mentioned have a greatest diameter of 0.5-0.7 mm, but there are also much smaller ones. The spines are about equally wide for a long stretch and then taper very gradually. The surface is smooth, or provided with faint longitudinal striae, as in Pl. 2, fig. 3, and Pl. 3, fig. 2. It may be that the spines were hollow as in the Echinoturidae (Jurassic

¹ In the Stockholm collections there is a Silurian boulder of brownish grey limestone from Norderstrand, N. of Visby, which is replete with needle-shaped spines, presumably of echinoids. They attain a length of up to about 12 mm with a diameter of up to 0.8 mm. They seem to be silicified, which is also true of certain other fossils in the same boulder. The stratigraphic horizon cannot be settled, but it is clear that the boulder does not originate from any part of the series of strata exposed on Gotland. It is likely to have been derived from some bed older than the Lower Visby marl and younger than the so-called Red bed which forms part of the rock-floor off the NW. coast of Gotland.

to Recent; see Mortensen 1935, pp. 80, 96) which, moreover, have been derived from the Paleozoic Lepidocentridae.

Spines similar to those now described are found e.g. in *Lepidocidaris* (Jackson 1912, Pl. 17, fig. 13) and *Lepidesthes* (Jackson 1912, Pl. 73, fig. 2). It is not necessary to say that this comparison does not involve any suggestion of relationship between the Silurian form (or forms) and the Carboniferous genera mentioned.

According to Hede (1942, p. 226 [22 of sep. print]), "the Slite group is Wenlockian in age not younger than the *Cyrtograptus ellesi* zone".

c. Specimens from the Hemse group

Pl. 3, fig. 4.

A small sample (RM Ec. 80) of so-called Etelhem reef-limestone, from Tänglings hällar (in the neighbourhood of Tänglings, about 1700 m south of the church of Etelhem) contains a fragment of the proximal portion of a minute spine. It has a length of 1.9 mm, and tapers from 0.7 mm to 0.5 mm. It is striated longitudinally (Pl. 3, fig. 4). In addition to this specimen there are a few other similar ones (RM Ec. 82a-b).

These specimens are essentially of the same type as the spines from the Slite group described in the preceding paragraph (for comparison, see Jackson's figures referred to above, and, further, his Pl. 19, fig. 5, showing spines of the Middle Devonian genus *Lepidocentrus*).

Another locality, Sandarve kulle, 1200–1300 m north of the church of Fardhem and approximately as far from (SW. of) Lind-klint, has yielded a somewhat different material. Several samples of greenish-grey reef-limestone (RM Ec. 78a–d, 79a–d) contain extremely slender (diameter 0.2 mm) smooth spines. It is conceivable that the material from Tänglings hällar and Sandarve kulle represent different species.

The stratigraphic horizon of these specimens is the same as that of *Silurocidaris clavata* n.sp. Possibly, one type may be secondary spines of this species.

C. Remains of Aristotle's lantern

In the material available, the jaw apparatus of echinoids is represented by quite a number of pieces integrated into that complicated structure known as Aristotle's lantern. As so often with old collections, many of the specimens are inadequately labelled. Only in a few cases is the locality stated definitely. This applies to RM Ec. 74, from Samsugn, Othem, and to two pyramids, from Lansa,¹ Island of Fårön, NE. of the main Island of Gotland. The specimens last mentioned (RM Ec. 84b 1–2) are accompanied by a small label (written by G. Holm?) on which is stated locality and derivation of the specimens, namely from "Fegraeus' Collection".² Other specimens (RM Ec. 84c–e, each number comprising several specimens) are marked "Gotland", without any further specification, others, finally, have the designation "Gotland?". However, the whole material is very uniform in state of preservation and colour. There is thus hardly any reason to doubt that all specimens originate from Gotland, and, in the writer's opinion, very likely from Lansa on the Island of Fårön. These specimens are completely weathered out of the rock and relatively very large-sized,

¹ On topographical map-sheets the name of this locality is written "Landsnäs" or "Landsnäsa".

² An extensive collection of fossils from Gotland brought together by L. Fegraeus, assistant land-surveyor, was purchased by the Swedish Museum of Natural History in 1879, partially by means of public funds (Holm 1916, p. 231).

while the specimens from Samsugn referred to above are embedded in the rock and relatively much smaller. In recognition of their difference in size and geological age (see below), the specimens from Samsugn and those from Lansa (or presumably from this locality) may represent different species and will be dealt with separately below.

Some students attach great importance to the lantern from a taxonomic point of view, and are of opinion that "the structure of its several parts presents characters that are of ordinal or subordinal value" (Jackson 1912, p. 177). This may be true. But where the Paleozoic forms are concerned there is a serious drawback: The material of Paleozoic — and particularly old Paleozoic — lanterns being strongly restricted, our information on the nature of the jaw apparatus is, on the whole, very incomplete, and entirely wanting in the case of several genera. By way of illustration, the conditions in the Palaeoecinidae were characterized by Jackson (1929, p. 37) as follows—.

"This remnant of an Aristotle's lantern [just described] in *Palaeoecinus ellipticus* is of much interest as the lantern in the whole family of the Palaeoecinidae is otherwise known only in *Melonechinus multiporus* (Norwood and Owen) ... and in part in *Oligoporus* and *Lovenechinus*." This statement is still valid. The character of the teeth is unknown (Mortensen 1935, p. 33).

The Lepidocentridae are comparatively better known, but very inadequately.

In the Archaeocidaridae, the lantern — or parts of it — has been described in several species of *Archaeocidaris*, indicating that the lantern varies a good deal in shape even within one and the same genus.

Unfortunately, the present writer has not been able to attribute the Gotland material with any degree of certainty to any of the major systematic units just mentioned, much less to an established genus. The reason for this will be discussed below. The logical consequence would be to leave the specimens without a name. On account of the uniqueness and the exquisite state of preservation of these remarkable lantern remains the author finally decided, however, to establish the specimens from Fårö with a specific name. Since a specific name cannot stand without reference to a higher systematic unit, it was an unavoidable necessity to create also a new genus in spite of all potential objections against such a step, the reasoning being much the same as in the case of *Silurocidaris* n. gen.

As stated above (p. 157), Lindström (1885, 1888) had already referred to the occurrence of lantern remains in the Silurian of Gotland.

Incerti ordinis and incertae familiae

Features of the dental apparatus considered to be essential for the characterization of the different orders within the Regularia (Endocyclica) have been plotted in Table 2, p. 171 (data mainly from Jackson 1912).

An examination of available data reveals that things are more complicated and less schematic than might be inferred from the table just referred to.

Let us first consider the character of the lantern, if erect or inclined. A complete lantern is only exceptionally available. However, if the pyramids be wide-angled in outline it follows as a corollary that the lantern will be inclined. In the Lepidocentridae there are forms which are not wide-angled and thus more or less erect (cf. text-fig. 3, nos. 6–7 but note that no. 6 should be more wide-angled). The depth of the foramen magnum, too, is subject to considerable variation in the Lepidocentridae. As is evident from text-fig. 3, it can be very shallow (nos. 3, 4, 7), contrary to what is demanded by the above scheme. In some orders, the tops of the pyramids have deep

Table 2.

Order	Lantern	Foramen magnum	Epiphyses	Pits in the top of pyramids	Styloid process
Melonechinoida Lepidocentroida	inclined	moderately deep	narrow	wanting	not extending above the base of the foramen magnum
Cidaroida	erect	very shallow	narrow	wanting	not extending
Aulodonta Stirodonta Camarodonta	erect (inclined)	deep	narrow, or else wide and uniting over the foramen magnum	present	extending above the base of the foramen magnum

pits which can be seen when the epiphyses are removed. According to the scheme given above, this feature does not occur in those orders which have Paleozoic representatives. However, in the species to be described below pits of this kind are obviously present. Accordingly, it should be excluded from the Melonechinoida, Lepidocentroida and Cidaroida but, on the other hand, the moderate depth of the foramen magnum, to say nothing of other characters, forbids association with orders considered entirely post-Paleozoic.

Thus the Gotland material cannot be forced into any of the established orders. In recognition of our still very fragmentary knowledge of the lantern in Paleozoic forms, it was thought wise to classify the genus to be erected below as *incerti ordinis* and *incertae familiae*.

Genus *Lanternarius* n. gen.

GENOTYPE.—*Lanternarius latens* n.sp.

DERIVATION OF NAME.—From Lat. *lanternarius*, lantern-bearer.

DIAGNOSIS (with reference to jaw-apparatus only).—A genus *incertae familiae* with large, or relatively large, pyramids, the ad-perradial (ad-interpyramidal) walls having aborally an internal excavation (or a system of excavations); epiphyses narrow, not uniting over the foramen magnum; foramen magnum moderately deep.

DISCUSSION.—The position of this genus in relation to other Paleozoic genera has already been elucidated to a certain degree in the preceding discussion. Particularly significant are the internal excavations in the ad-perradial (interpyramidal) walls of the pyramids.

Text-fig. 3 was prepared to give a general idea of the outline of the pyramids in all previously recorded Upper Silurian–Lower Carboniferous genera better known in this respect. *Lanternarius* n. gen. seems in the first place to recall *Archaeocidaris* (angle of pyramid 45° – 50° in both; the relative depth of the foramen magnum somewhat greater in *Lanternarius*), and some genera among the Lepidocentridae. But it would be premature to interpret this similarity as an expression of affinity; it may be merely casual.

Another object of the text-fig. mentioned was to demonstrate eventual trends in the phylogenetic development of the lantern (or, more precisely, the pyramids), and, if






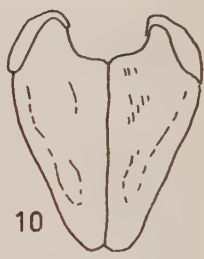


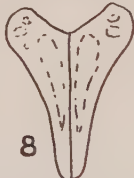

	Palaeechinidae	Lepidocentridae	Archaeocidaridae
L. CARB.	 1  2	 3  4  5	 10
U. DEV.		 6	
M. DEV.		 7	
U. SIL.		 8  9	

Fig. 3. Pyramids of previously described Upper Silurian–Lower Carboniferous echinoids, arranged stratigraphically. In genera not recorded in this table, the lantern is either unknown, or poorly known. 1, *Melonechinus multiporus* (after Jackson 1912, Pl. 56, fig. 9); 2, *Palaeechinus ellipticus* (after Jackson 1929, Pl. 2, fig. 16); 3, *Pholidechinus brauni* (after Jackson 1912, fig. 207); 4, *Lepidesthes wortheni* (after Jackson 1912, Pl. 67, fig. 12); 5, *Meekechinus herborensis* (after Bindemann 1938, Pl. 1, fig. 1a); 6, *Devonocidaris jacksoni* (after Thomas 1920, Pl. 51, fig. 5; p. 542: “restored ...; the foramen magnum should be wider”); 7, *Lepidechinoides hunti* (after Cooper 1931, fig. 2A); 8, *Echinocystites pomum* (after Gregory 1897, fig. 3); 9, *Palaeodiscus ferox* (combined after Hawkins & Hampton 1927, fig. 6B, and MacBride & Spencer 1939, fig. 11 C); 10, *Archaeocidaris rossica* (after Jackson 1912, fig. 208). All external faces, save no. 8. Not to scale.

such could be recognized, to fix the place of *Lanternarius* in a line of evolution. Unfortunately, however, we must accept the fact that the material available is basically insufficient to give any evidence in either respect.

REGIONAL DISTRIBUTION.—Sweden (Gotland).

STRATIGRAPHIC RANGE.—Middle Silurian.

Lanternarius latens n. gen. n. sp.

Pl. 2, figs. 4–6. Text-fig. 4.

DERIVATION OF NAME.—From Lat. *latens*, lying concealed, unknown, in allusion to the fact that no parts of this species are known except pieces of the lantern.

HOLOTYPE.—RM Ec. 84b1.

TYPE LOCALITY.—Lansa, Island of Fårön, Gotland.

TYPE STRATUM.—Judging from scraps of rock adhering to the holotype, this was contained in a grey, marly limestone. According to kind information from Dr. J. E. Hede, the fossils from Lansa most probably originate from the very lowest member of the Slite group, although the topmost Högklint group (the Tofta limestone is not developed in N. Gotland) cannot be left out of account entirely. However that may be, the type stratum is of Wenlock age.

MATERIAL.—The holotype is a well-preserved pyramid though somewhat worn, with remains of associated parts. Along with it, there is another, more fragmentary pyramid (RM Ec. 84b2). Concerning the rest of the material assigned to this species, see also p. 169 above. It includes one not very well-preserved pyramid (RM Ec. 84a1), 18 right maxillae (RM Ec. 84a10–16, 84c1–4, 84e1–7), all more or less fragmentary, and 12 left maxillae (RM Ec. 84a2–9, 84d1–4), also more or less fragmentary.¹

DIAGNOSIS.—Same as that of the genus, by monotypy.

DESCRIPTION.—It will be convenient to describe the figured specimens which are those best preserved and, where eligible, make comments on the specimens not figured.

The holotype (Pl. 2, figs. 6a–b; text-fig. 4: 1) measures about 19 mm from the lateral wings (inclusive) to the adoral margin of the pyramid. The greatest width, which is on the same level as the adoral margin of the foramen magnum, is about 13 mm. The greatest depth of the foramen magnum corresponds to approximately 25 per cent of the total height of the pyramid. The pyramid angle is about 45°. The outer surface is smoothly convex along the pyramidal suture. The external margin is sharp, merging gradually into the adoral tips of the maxillae. The admedian plane is vaulted, being separated from the margin by a deep sulcus. The left maxilla carries a narrow epiphysis on its shoulder. The inner face of the pyramid shows the somewhat damaged interpyramidal surfaces, forming an angle of about 90° with each other. The corrugation is but slightly visible in this specimen. At the aboral end of the pyramid there are fragments of some other elements of the lantern skeleton. The piece in the upper left corner of Pl. 2, fig. 6b, will be a rotula, whereas that in the upper right corner is likely to be part of an epiphysis. The tooth is not preserved.

The specimen RM Ec. 84b2 is essentially similar to the holotype. A detail that is visible in this specimen but not in the holotype is that the margins of the maxillae meeting in the pyramidal suture are serrate, with six notches to 1 mm.

The third pyramid available, RM Ec. 84a1, is much smaller, measuring 11 mm in height, and 5.5 mm in greatest width. The minor relative width may at least partially be due to the fact that the outer margins are not very well preserved in this specimen. Transverse ridges on the interpyramidal surfaces are well visible. An aboral part of

¹ It should be mentioned that specimens RM Ec. 84a are accompanied by a label (written by G. Holm?) the text of which runs as follows: "*Paleechinus siluricus* ANGELIN in Msc. Tafl. VIII figg. 10–16. Gotland??" Whether or not the manuscript referred to is still in existence the present writer does not know, nor has he seen any figures of Silurian echinoids on unpublished plates by Angelin preserved in the Paleozoological Department of the Swedish Museum of Natural History. The same name is written in pencil on the lower surface of the small round boxes with glass-lids containing all specimens save 84a.

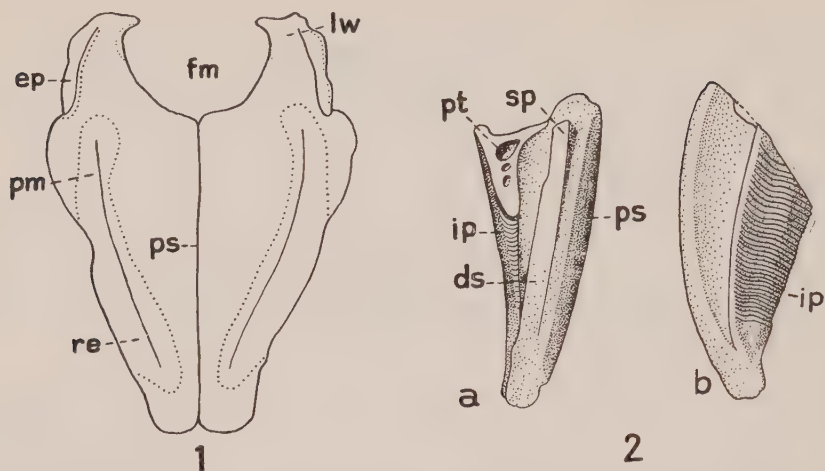


Fig. 4. *Lanternarius latens* n.gen. n.sp., parts of Aristotle's lantern. 1, pyramid, external face; 2a, right maxilla, internal face; 2b, the same, interpyramidal face. *ds*, dental slide; *ep*, epiphysis; *fm*, foramen magnum; *ip*, interpyramidal suture; *lw*, lateral wing; *pm*, protractor muscles; *ps*, pyramidal suture; *pt*, pits in upper part of half pyramid; *re*, retractor muscles; *sp*, styloid process. $\times 3$. — Sweden, Gotland.

the tooth is preserved. It seems to be grooved, but unfortunately the state of preservation is not as perfect as to admit of a definite statement.

The right maxilla RM Ec. 84e1 (Pl. 2, figs. 5a–b; text-fig. 4: 2) is incomplete in that the aboral portion is wanting. The greatest width of the interpyramidal joint-face is presumably at the aboral end. The transverse ridges for attachment of interpyramidal muscles have a very slight S-curve but run, on the whole, perpendicularly to the long axis of the surface. Aborally, there are 5 ridges to 1 mm, adorally 6 ones. The surface is slightly concave. The outer surface of the maxilla has largely the same characters as that of the holotype, but as in several other free maxillae, the admedian area is shaped like a rounded ridge. The internal face is narrow. In the central canal is lodged the dental slide which is produced aborally into a short styloid process. In this specimen, and in a number of other ones, deep pits are seen to be present in the ad-interpyramidal wall (see text-fig. 4: 2a, and Pl. 2, fig. 5a).

The left maxilla RM Ec. 84d1 (Pl. 2, fig. 4) does not require much comment, being an image of the right maxilla. The symphyseal face bordering on the pyramidal suture is narrow, being somewhat canoe-like in outline. Measurements of the maxillae were not given above since all specimens available are imperfect in one respect or another.

DISCUSSION.—Little can be added to what has been said above about the relationships of *Lanternarius* to other genera. Judging from the size of the pyramids, which attain a height of 19 mm, the lantern was extraordinarily large. For comparison, the following data on the size of the pyramids were compiled from different sources:

Height of pyramids	> 10 mm	in 7 species;
" "	10–15 "	" 6 "
" "	15–17 "	" 2 "

From the size of the lantern it may be inferred, moreover, that the corona of *Lanternarius* was of considerable size. To give a rough idea of its size it may be mentioned

that in a specimen of *Echinus esculentus* (Recent) the diameter of the corona is 78 mm, corresponding to a pyramid height of 22 mm. Provided the proportions of the height of the pyramids to the diameter of the corona were the same in *Lanternarius* (which is of course an unconfirmed assumption), the diameter of the corona would be about 67 mm in the holotype.

It is obvious that the lantern in *Lanternarius* was rather different in shape from that in the Silurian genera *Echinocystites* and *Palaeodiscus*, as the pyramids do not conform in outline to each other in the genera mentioned (cf. text-fig. 3, nos 8–9, and text-fig. 4: 1). These are the only Silurian forms in which the dental apparatus has been figured so far. But, according to Jackson (1912, p. 236), "an excellent echinoid lantern from the Wenlock Limestone of Dudley" is present in the collections of the British Museum (Nat. Hist.). This lantern was described by Jackson (1912, p. 236, footnote 1) as follows: "It has wide-angled pyramids, a moderately deep foramen magnum, and a brace [rotula] in place. The sides of the pyramids have ridges for the attachment of interpyramidal muscles. It has the essential characters of lanterns, as seen in the Lower Carboniferous. As there is no portion of the test in association with the lantern, its generic position is unknown, but it is quite probable that it is a *Lepidocentrus* or a near ally of that genus." The description is too generalized to admit of a comparison with the lantern parts from Gotland. About the attribution to *Lepidocentrus*, cf. above p. 156, foot-note 2.

REGIONAL DISTRIBUTION.—Sweden, Gotland, Fårön: Lansa (the derivation of many specimens assigned to *Lanternarius latens* is not known with certainty, as observed above, p. 169).

STRATIGRAPHIC RANGE.—Middle Silurian, Wenlock (about the age of the Slite group cf. above p. 169).

Gen. et sp. indet.

Pl. 2, fig. 3.

These fragments of a small lantern (right maxillae, the largest one having a height of 7 mm) are interesting inasmuch as they are associated with fragments of spiniform radioles (cf. above p. 168). These parts are very likely conspecific. This, however, does not help greatly, because the spine cannot be determined.

As previously mentioned, the species represented by these insignificant fragments, the characters of which cannot be ascertained, has not been considered conspecific with *Lanternarius latens*, on account of its much smaller size (this feature is, of course, not decisive), although the stratigraphic horizon is more or less the same (the Slite group). The locality is Samsugn, parish of Othem.

4. General remarks

As will have appeared from the preceding pages, the echinoid fauna of the Silurian of Gotland — as represented by the fossil material — is neither abundant, nor varied. Yet, by the extreme rarity of Silurian echinoid remains in the world fossil record, the Gotland echinoids can lay claim to a considerable interest.

Their stratigraphic range and geographic occurrence are summarized in Table 3, p. 176.

The table shows that echinoid remains are scattered throughout the series of strata from the Upper Visby marl (inclusive) to the Hemse group (inclusive), of Valentian (Llandovery) to Lower Ludlow age. The finds are few, however, and the number of individuals small. This can very likely be explained by a primary scarcity of these

Table 3. Range and localities of Silurian echinoids so far known from Gotland.

	Va- len- tian	Wenlock						Lud- low								
	Upper Visby marl	Höglint group	Tofta limestone	Slite group	Halla limestone	Mulde marl	Klinteberg group	Hemse group	Vattenfallet (Visby)	Lansa (Fårön)	Samsugn (Othem)	Klinteberget	Lind-klint	Manngårde (Lye)	Tånglångs hållar (Etelhem)	Sandarve kulle (Fardhem)
<i>Gotlandechinus balticus</i> n. sp.	+	+
<i>Silurocidaris clavata</i> n. sp.	+	+	.	.	.
Spiniform radioles	+	+	.	+	.	.	.	+	+	.	+	.	.	.	+	+
<i>Lanternarius latens</i> n. sp.	+	+
Jaw-fragments, gen. et sp. indet.	+	+

forms which seem to have played a subordinate role in the biocoenoses on the bottom of the sea which covered Gotland in Silurian times. Otherwise, their remains could reasonably be expected to be as abundant as, for example, those of the crinoids. There are several gaps in the stratigraphic distribution. Those in the Tofta limestone and the Mulde marl may possibly be due to ecological conditions unfavourable to the vigour of the echinoids. As we have seen, the Gotland echinoids seem to have lived mainly on reefs, whereas reef-facies did not prevail in the area during the deposition of the beds that form the Tofta limestone and the Mulde marl.

Since all forms treated in the present paper have had to be assigned to new genera (the spiniform radioles could not be determined generically) which are clearly distinct from all — though these are indeed very few — old Paleozoic echinoids previously described, it is at present impossible to express an opinion on the interrelationships between this and foreign echinoid faunas of the Silurian, their possible centre of evolution and dispersal, etc.

It has been considered, up till now, that the Melonechinoida, with the single family Palaeoecchinidae, were confined to Lower Carboniferous deposits of Europe and N. America. By the introduction of the new family Gotlandechinidae, with *Gotlandechinus* n. gen., the geologic history of the Melonechinoida — leaving the somewhat doubtful *Maccoya phillipsiae* (FORBES) (cf. p. 156) out of account — has now been prolonged some 60–70 millions of years backwards in time. There is thus no longer any reason to look upon the Melonechinoida as a rootless assemblage with a short span of life (see e.g. Beurlen 1937, p. 20), but rather as a slowly developing branch “exploding” into a diversity of branchlets in Eocarboniferous times. As indicated above (p. 164), Termier & Termier (1950, Pl. 232, fig. 5; not mentioned in the text) figured a thin-section of a spine of *Palaeoecchinus* from the Devonian (Eifelian) of Morocco. Supposing that the attribution to *Palaeoecchinus* is correct which can hardly be demonstrated conclusively this find contributes to bridge the gap between the Silurian and the Carboniferous Melonechinoida.

With regard to *Silurocidaris* n. gen. it enables us to trace the evolutionary line of the Cidaroida backwards from the Devonian (*Xenocidaris*, *Nortonechinus*) into the Silurian.

From a phylogenetic point of view, it is an interesting fact that certain features in the organization of late Paleozoic and younger echinoids were evolved in close agreement with an archaic pattern. This is true of the lantern, the structure of which has changed amazingly little in the long span of time from the dawn of the Paleozoic up to Recent times. That this is so was observed already by Jackson (1912, p. 182), Thomas (1920, p. 482), and Hawkins (1943, p. LVII). The latter remarks that the lantern "seems to have attained virtual perfection in Silurian times". The evolution of the lantern was commented upon by Sollas (1899, p. 710 *et seq.*) and was discussed thoroughly by Jackson (1912, p. 177 *et seq.*).

The significance of the coronal structure in *Gotlandechinus* was touched upon above in discussing the genus and the species attributed to it. It is the writer's hope to have an opportunity in another connexion to deal, from a more general point of view, with problems of echinoid phylogeny raised by early forms.

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Tryckt den 6 december 1956

Uppsala 1956. Almqvist & Wiksells Boktryckeri AB

PLATES

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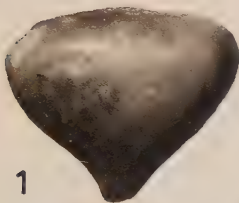


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- (Phot. U. Samuelsson. Ret. S. Ekblom).



3



1



4



5a



5b



6a



2



6b

Plate III

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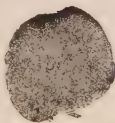
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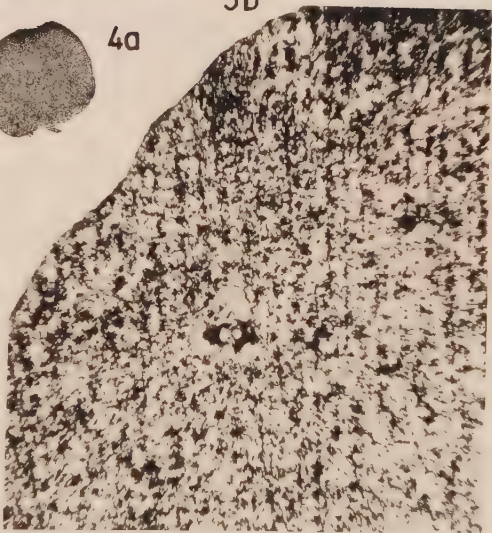


Plate IV

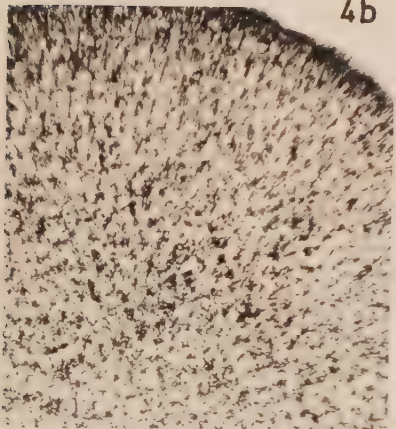
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5b



4b



A preliminary report on the structure of the siphonal tube and on the precipitation of lime in the shells of fossil Nautiloids

By HARRY MUTVEI

With 1 plate and 4 figures in the text

Introduction

The present paper communicates some of the results of the writer's recent investigations into the shell structure of Paleozoic Nautiloids. In order to explain the structure of the siphonal tube in these Nautiloids a general survey of the structure of the siphonal tube in *Nautilus* and *Spirula* is also included. This latter survey is based, in part, on new investigations carried out by the writer.

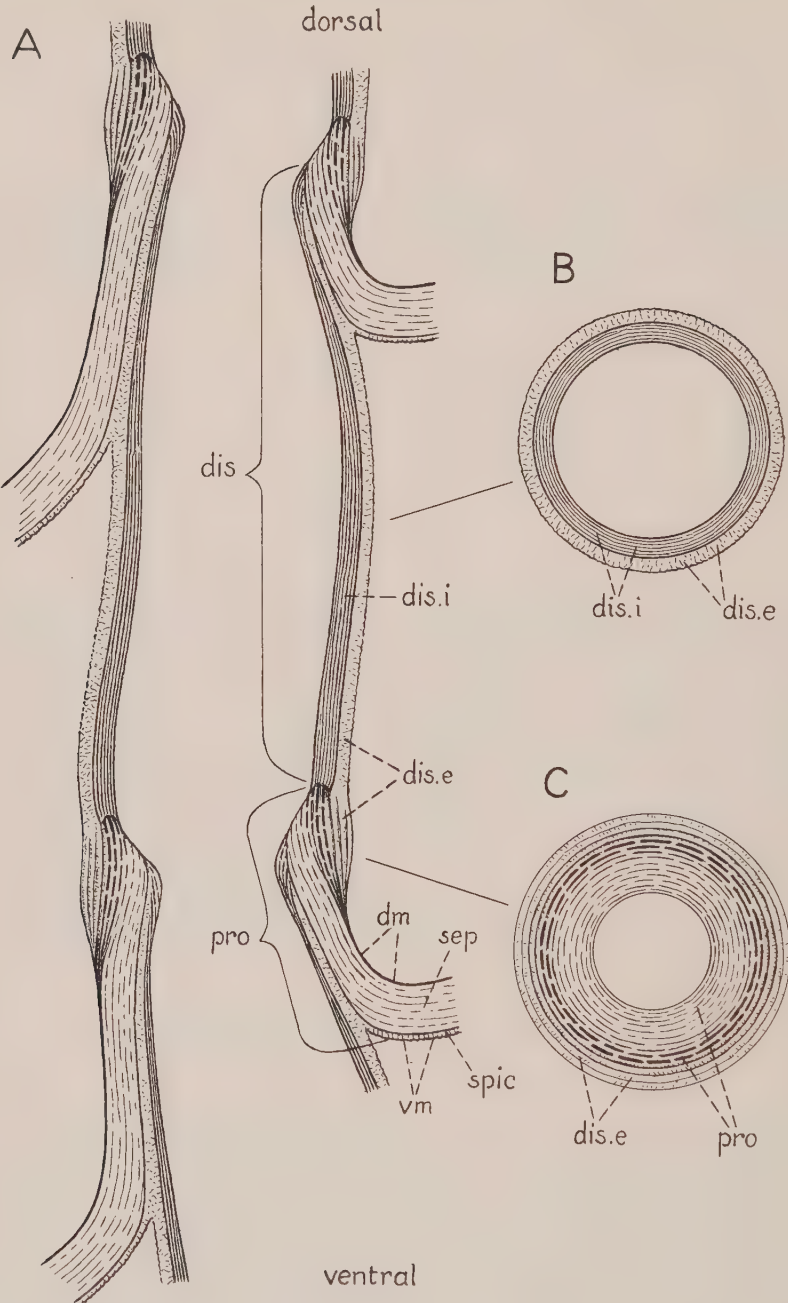
Many previous writers (Huxley 1853, Balfour 1880, Hescheler 1900, Pelseneer 1906, Plate 1922, Naef 1926, and Mutvei 1956) have pointed out that the Cephalopod body must be orientated on the basis of comparative anatomy and with due regard to conditions in molluscs in general. Thus the head and the funnel then lie ventral and the dilated portion of the mantle cavity, with the pallial organ-complex, postero-ventral. Since the shell is secreted by the epithelium on the dorsal surface of the body proper, it lies, morphologically, with the aperture of the shell directed downwards and its apex upwards. In the following description the shell is orientated in this way.

The structure of the septum in *Nautilus* and *Spirula*

a) The septum proper

It is well known that two main divisions can be distinguished in each individual septum of *Nautilus* and *Spirula*: (a) the *septum proper*, and (b) the *siphonal funnel* ("septal segment", Flower).

The septum proper is, in *Nautilus*, composed of the following four layers: (a) a dorsal chitinous membrane (*dm*, Fig. 1 A; Appellöf 1893, *mb*, pl. 11:1; 12:1); (b) a nacreous layer, made up of numerous lamellae (*sep*, Fig. 1 A; Appellöf loc. cit., *sp*, pl. 11:1; 12:1); (c) a thin layer of needle-shaped calcareous elements, the *spicular layer* (*spic*, Fig. 1 A; Appellöf loc. cit., *pf*, pl. 12:1, 3); and (d) a ventral chitinous membrane (*vm*, Fig. 1 A; Appellöf loc. cit., *mb*, pl. 12:3). The spicular layer is fairly thick in the



proximity of the siphonal funnel, but probably thins out and disappears towards the shell wall.

In *Spirula* the septum proper has a simpler structure than in *Nautilus*; it is composed only of a nacreous layer (*sep*, Fig. 2 A; Appellöf loc. cit., *sp*, pl. 8:1; 9:1, 2, 5, 6) and a spicular layer; the latter is, however, to be found only in close proximity to the siphonal funnel (*spic*, Fig. 2 A, C; Appellöf loc. cit., *pf*, pl. 8:1; *d*, pl. 9:1). According to the writer's own observations the spicular layer in *Spirula* is not continuous but is represented by radiating ridges (*spic*, Fig. 2 C).

b) The siphonal funnel

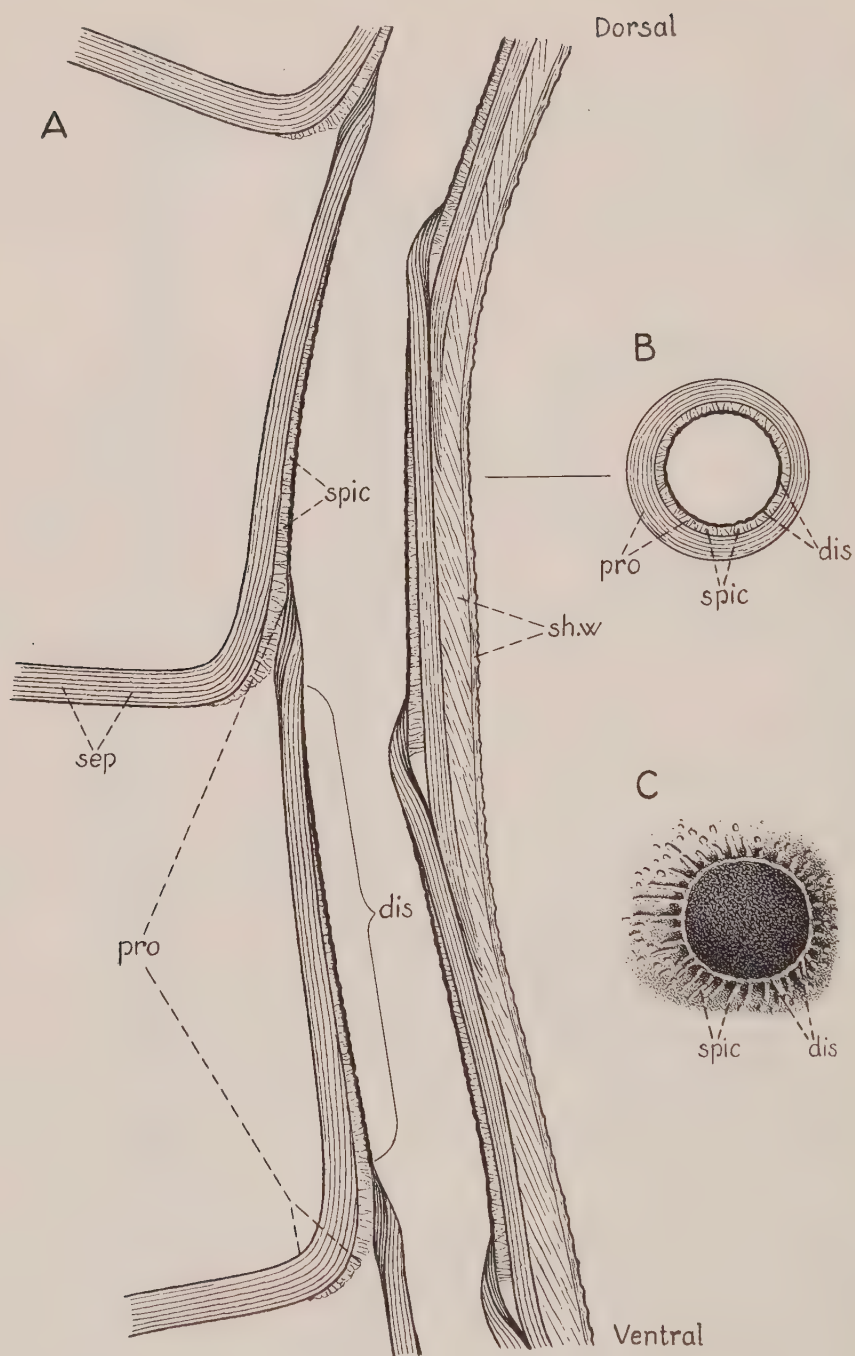
The septum proper in *Nautilus* and *Spirula* is produced upwards into a siphonal funnel, which extends to the next older siphonal funnel, to the inside of which it is attached (Barrande 1877 a, Brooks 1888, Appellöf loc. cit.). Consequently the siphonal funnels form a continuous tube. This tube, the *siphonal tube* ("siphuncle", Owen, Hyatt; "siphon", Barrande, Griffin; "Sifo", Appellöf; etc.), encloses the *siphonal cord* ("siphuncle", Griffin, Willey; "siphon", Owen, Hyatt; "fleischiger Sifo", Appellöf; etc.), i.e. the dorsal, narrow portion of the body extending up towards the shell apex. In the siphonal funnel of both forms one can distinguish two structurally different divisions: (a) a *proximal* (*pro*, Figs. 1, 2; "funnel", Hyatt, Brooks; "goulot", Barrande; "Siphonaldute", Sandberger, Appellöf; etc.); and (b) a *distal* (*dis*, Figs. 1, 2; "sheath", Hyatt; "enveloppes siphonales", Barrande; "Siphonalhülle", Appellöf; "connecting ring", Flower; etc.).

With regard to its structure, the proximal division in *Spirula* is similar to the septum proper, i.e. it is composed of two layers, the nacreous layer and the spicular layer (*pro*, Fig. 2 A, B; Appellöf loc. cit., *dt*, *pf*, pl. 8:1, 3; 9:5). In *Nautilus* this division is made up of three of the four layers present in the septum proper, or, more precisely, the dorsal chitinous membrane, the nacreous layer, and the spicular layer (*pro*, Fig. 1; Appellöf loc. cit., pl. 12:1), whilst the ventral chitinous membrane is lacking. It may also be noted that the proximal division has a considerably greater height in *Spirula* than in *Nautilus*. Its height is, in the former, as much as $\frac{3}{8}$ and in the latter as much as $\frac{1}{4}$ to $\frac{1}{3}$ of the total height of the siphonal funnel.

The distal division in *Nautilus* is composed of two layers, (a) an external, porous, calcareous layer, which is largely made up of small, needle-shaped calcareous elements (*dis.e*, Fig. 1 A, B, C; Appellöf loc. cit., *pf*, pl. 11:2, 4; 12:1; Brooks 1888, *a*, *ax*, pl. 1, 2); and (b) an internal chitinous layer (*dis.i*, Fig. 1 A, B); the latter contracts on drying and appears, therefore, in the dried shells as a chitinous membrane only (Appellöf loc. cit., *chm*, pl. 11:2, 3; 12:1; Brooks loc. cit., *f*, *fx*, pl. 1, 2). In *Spirula*, on the contrary, the division under discussion is composed of a thin calcareous layer only (*dis*, Fig. 2 A, B, C; Appellöf loc. cit., *hü*, pl. 8:1, 3), made up of small, rounded calcareous

Fig. 1. Diagram to show the structure of the siphonal tube in *Nautilus pompilius*, L. from several thin-sections of a shell preserved in alcohol and plastic embedded. A, lower portion of the siphonal tube in median-sagittal section; B, C, cross-section through the distal division of the siphonal funnel and the upper portion of the proximal division. $\times 8$.

dis, distal division of the siphonal funnel; *dis.e*, external calcareous layer of the distal division; *dis.i*, internal chitinous layer of the distal division; *dm*, dorsal chitinous membrane of the septum proper; *pro*, proximal division of the siphonal funnel; *sep*, nacreous layer of the septum proper; *spic*, spicular layer of the septum proper; *vm*, ventral chitinous membrane of the septum proper.



éléments, and which in *Nautilus* corresponds to the external calcareous layer of the distal division. The internal chitinous layer of the distal division in *Nautilus* is, therefore, entirely lacking in *Spirula*.

The two divisions of the siphonal funnel in *Nautilus* are connected with each other in such a way that the external calcareous layer of the distal division encloses the upper half of the proximal division and merges into it without any distinct boundary (Fig. 1 A; Appellöf loc. cit., *ds*, pl. 12:1; Brooks loc. cit., pl. 1), whilst the internal chitinous layer of the distal division has, on the contrary, a clearly defined boundary at the upper margin of the proximal division (Fig. 1 A). In *Spirula* the distal division of the siphonal funnel is connected with the upper portion of the proximal division (Fig. 2 A; Appellöf loc. cit., pl. 8:1) in a manner similar to that of the external calcareous layer of the distal division in *Nautilus* just described.

The individual siphonal funnels in *Nautilus* are joined in such a way that the upper half of the distal division in each siphonal funnel extends upwards into the next older one and invests the inside of the proximal division of it. The external calcareous layer of the distal division therefore connects with, and merges into, the spicular layer of the next older septum (Fig. 1 A; Appellöf loc. cit., pl. 12:1; Brooks loc. cit., pl. 1, 2). In *Spirula* the individual siphonal funnels join each other, in principally the same way as in *Nautilus*. Owing to the fact that the proximal division of the siphonal funnel in this form is very high, the entire distal division invests the proximal division of the next older siphonal funnel (Fig. 2 A; Appellöf loc. cit., pl. 8:1).

The structure of the siphonal tube in fossil Nautiloids

The siphonal tube in all fossil Nautiloids, as in *Nautilus* and *Spirula*, is composed of successive siphonal funnels, each of which is composed of two structurally different divisions, a proximal and a distal. Owing to the fact that the fossil Nautiloid-shells are always more or less recrystallized the siphonal funnel shows little or nothing of its original structure (Barrande 1877 a).

It is well known that the two divisions of the siphonal funnel vary considerably in height in different fossil Nautiloids. The height of these divisions in many fossil forms (e.g. *Orthoceras*, *Michelinoceras*, *Estonioceras*) corresponds to those in *Nautilus*, whilst some other fossil forms (e.g. *Endoceras*, *Piloceras*, *Aturia*) resemble *Spirula* in this respect. Moreover, the distal division of the siphonal funnel varies in thickness (Holm 1899; Flower 1941 a, 1943 b, 1946, 1947, 1950; Ulrich, Foerste, Miller & Unklesbay 1944; Cecioni 1953; Teichert & Glenister 1954), but the variations in this character are considerably smaller than has been previously stated. According to Holm (loc. cit.) this division of the siphonal funnel in *Bathmoceras* is so thick that it forms a very prominent ridge on the inside of the siphonal tube. *Bathmoceras* would in this respect differ from all remaining Nautiloids. It is, however, quite clear that this ridge does not belong to the division under discussion, but is a later secondary thickening on the

Fig. 2. Diagram to show the structure of the siphonal tube in *Spirula spirula* (L), from several thin-sections of shells preserved in dried condition and plastic embedded. A, lower portion of the siphonal tube in median-sagittal section; B, cross-section of the siphonal tube; C, spicular layer, represented by radiating ridges, at the passage between the septum proper and the siphonal funnel; the distal division of the next older siphonal funnel (*dis*) broken off. $\times 28$.

dis, distal division of the siphonal funnel; *pro*, proximal division of the siphonal funnel; *sep*, calcareous layer of the septum proper; *sh.w.*, shell wall; *spic*, spicular layer of the septum proper.

inside of the siphonal tube (see p. 186). Again in *Eothisnoceras* (Ulrich, Foerste, Miller & Unklesbay loc. cit.; Teichert & Glenister loc. cit.) and in *Cyrtocercina* (Flower 1943 b, 1946) the inside of the distal division in every siphonal funnel would form a prominent ridge, but in the opinion of the present writer this ridge is also probably of the same nature as in *Bathmoceras*, i.e. a later thickening on the inside of the siphonal tube.

Flower (1939 b, 1941 a, 1946, 1947) states that the distal division of the siphonal funnel is, in all Nautiloids, a mesodermal structure. This interpretation is quite incompatible with an anatomical point of view. Further, according to Flower (1941 a, b, 1943 b, 1946, 1947, 1950), the upper portion of the distal division, the "eyelet", has in certain fossil Nautiloids, e.g. *Tarphyceratidae* and *Endoceratidae*, a different structure from the rest of this division. It is, however, very doubtful if this is actually the case. To judge from Flower's figures the shells of the illustrated Nautiloids are strongly recrystallized and it could, therefore, be difficult to establish what the original structure of the siphonal funnel really was. The writer's close study of the structure of the siphonal funnel in a large, and well preserved, collection of material, e.g. *Endoceratidae*, did not confirm Flower's observations.

It is well known that the septum proper in all Nautiloids is secreted by the epithelium on the dorsal surface of the body proper. The siphonal funnel, on the contrary, is formed by the epithelium of the siphonal cord, or more exactly, on the newly formed part which results from the elongation of the siphonal cord. After having formed the siphonal tube the epithelium of the siphonal cord generally ceased entirely the secretion of shell substance. In certain Paleozoic Nautiloids (e.g. *Endoceratidae*, *Piloceratidae*, *Actinoceratidae*, *Trimeroceratidae*, etc.) the epithelium on the upper portions of the siphonal cord began, however, after an interval to secrete again. Thus there arose in these forms a new calcareous deposit on the inside of the siphonal tube, the *secondary endosiphonal deposit* ("dépôt organique dans le siphon", Barrande; "endosiphonal deposit", Hyatt; "endosiphuncular deposit", Ruedemann; "endosiphonal stereoplasmic deposit", Kobayashi; "siphonal deposit", Flower). This appears in three different types: (a) *conical* ("dépôt conique", Barrande); (b) *radial* ("actinosiphonate deposit", Flower); and (c) *annular* ("annulosiphonate deposit", Flower). It must therefore be presumed that the secondarily lime-secreting epithelium was differentiated in three ways. The conical endosiphonal deposit (Fig. 3 C), which from my findings is a continuous layer, was in all probability secreted by a horizontal epithelial zone, the height of which corresponded to several siphonal funnels. The radial endosiphonal deposit (Fig. 3 A) is composed of a great number of vertical, radially arranged, lamellar cristae ("lamelles rayonnantes", Barrande; "rays", "longitudinal lamellae", Flower; "radiating laminae", "internal calcareous septa", "radiating lamellae", "actiniform lamellae", Hyatt; etc.). These must have been formed by corresponding vertical epithelial bands (Barrande 1855, 1877 b), which probably extended through at least one siphonal funnel. The annular endosiphonal deposit (Fig. 3 B) is made up of a series of horizontal ring-shaped ridges, the *annular ridges* ("anneaux obstruteurs", Barrande; "rosettes", "rings", Hyatt; "annuli", Foerste, Teichert, Flower; "bulettes", Strand; "obstruction rings", Kobayashi, Teichert; etc.), one of which is present on the inside of each siphonal funnel. The epithelium which secreted these ridges must therefore have been composed of a small number of horizontal zones, one to each ridge (Barrande 1855, 1877 b).

Due to the continued lime-secretion the three types of secondary endosiphonal deposits grew in thickness, and consequently the cavity of the siphonal tube was correspondingly reduced which brought about a similar decrease in the diameter of the

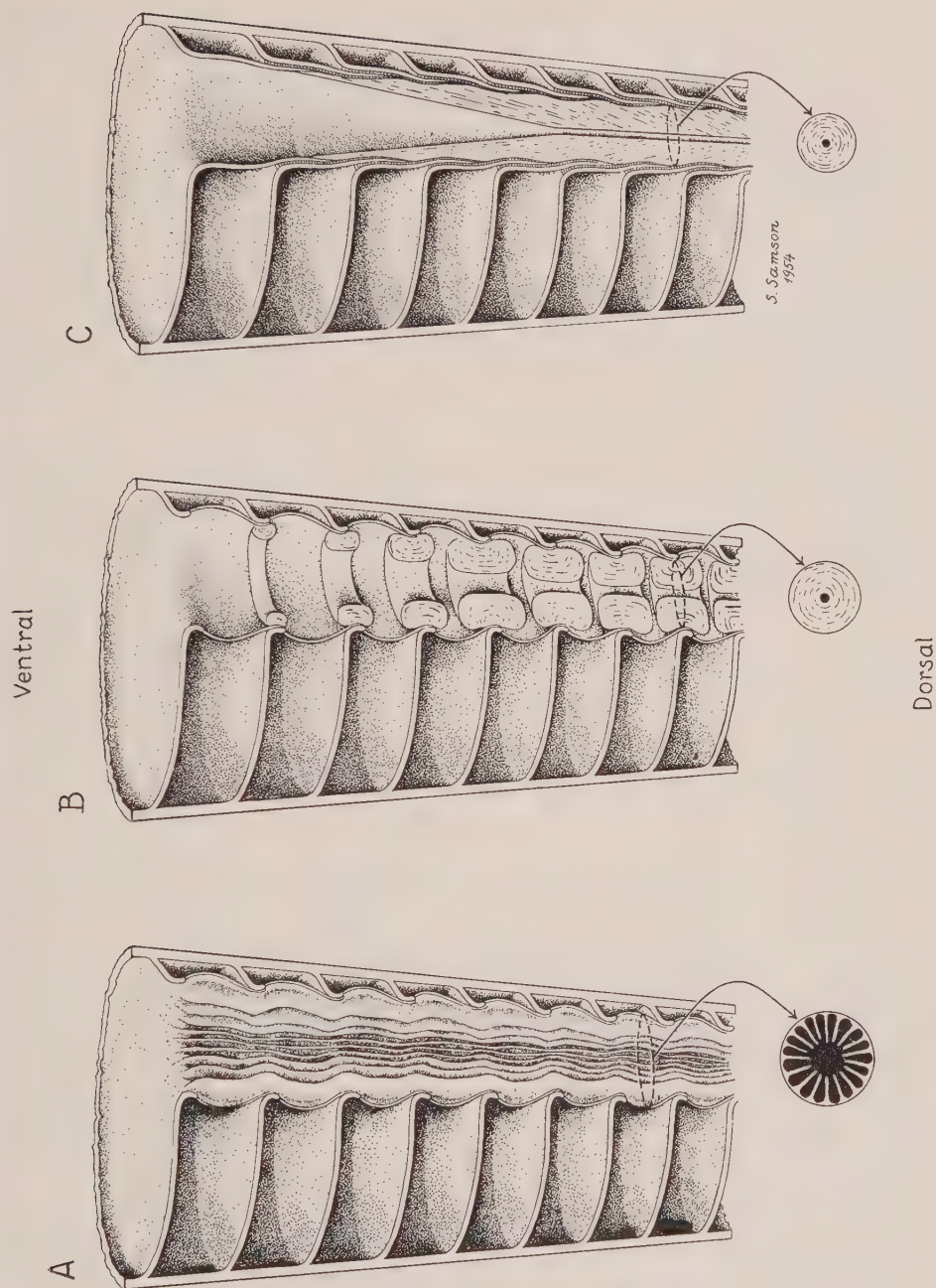


Fig. 3. Schematic representation of the three types of the secondary endosiphonal deposits. *A*, radial; *B*, annular; and *C*, conical.

siphonal cord in the locality of the lime secreting epithelium (Barrande 1855, 1877 b). The siphonal cord in the conical endosiphonal deposit became, therefore, distinctly narrower along its whole height (Fig. 4 C), whilst the siphonal cord internal to the annular and the radial endosiphonal deposits showed deep depressions, which in the former case were horizontal (Fig. 4 B; Barrande 1855, pl. 6:10-14) and in the latter case vertical (Fig. 4 A).

The secondarily lime-secreting region of the epithelium of the siphonal cord arose, in my opinion, on the upper portions of the siphonal cord and then moved successively downwards during the growth of the animal. In certain cases the movement continued as far as the lowermost portion of the siphonal cord in the full-grown animal. This downward movement occurred in such a way that the epithelium below the lime-secreting region regained the capacity for secreting lime, whilst the upper portion of this region at the same time lost this function. Simultaneously the endosiphonal deposit grew downwards, either by the continuous secretion of lime on its lower margin as in the conical and radial types, or by the upgrowth of new ring-shaped ridges below the next older one, as in the annular type.

With regard to its structure, the conical endosiphonal deposit agrees with the septum proper. When the conical endosiphonal deposit is only slightly recrystallized it has distinct growth lamellae which have arisen through smaller variations in the composition of the secretion. These growth lamellae have been misinterpreted by all previous writers (e.g. Barrande 1855, 1877 b; Hyatt 1883, 1900; Holm 1885, 1895; Ruedemann 1903; Kobayashi 1934, 1936 b, 1947; Flower 1941 a, 1950, 1955 b) as a series of separate calcareous walls ("gaines coniques", Barrande; "sheaths", Hyatt; "endocones", Hyatt, Ruedemann, Foerste, Holm, Flower; "endosiphosheaths", Ruedemann; "endosipholining", Ruedemann, Flower; "holoendosheaths", "endosiphuncular endosheaths", "interendosheaths", "subsheaths", Kobayashi; "Endosiphonalscheiden", Schindewolf; etc.), which according to Holm and Ruedemann would be attached to each other by means of a small number of radially arranged lamellae, called by Holm "endosiphoblades". To the number and position of these "endosiphoblades", Flower (1950) attaches great taxonomic value. From my own investigations it is evident that the "endosiphoblades", described by Holm, are only vertical fissures in the endosiphonal deposits, formed after the death of the animals. In all probability most of the "endosiphoblades", which Ruedemann and Flower have described, are similar post mortem fissures.

As a rule, every individual annular ridge in the annular endosiphonal deposit arose on the inside of the proximal division of the siphonal funnel (Barrande 1855, 1877 b; Hyatt 1900; Teichert 1933). An exception is, however, constituted by some few forms (*Bathmoceras* and probably also *Eothinoceras* and *Cyrtocerina*), in which every such ridge was, instead, formed on the inside of the distal division of the siphonal funnel. In the former case the annular ridges have the same structure as the septum proper, whilst in the latter case, on the contrary, they correspond structurally to the distal division of the siphonal funnel.

The structure of the radial endosiphonal deposit is at present unknown.

The origin of the endosiphonal deposit was discussed by Teichert (1933, 1935) and Flower (1941 a, 1942, 1943 b, 1946, 1955 b) who both arrived at conclusions which are quite erroneous from an anatomical point of view. Teichert states that the annular endosiphonal deposit was formed by a calcification of the tissues of the siphonal cord around an "endosiphuncular vascular system" in the siphonal cord. According to Flower all endosiphonal deposits were formed in the mesoderm.

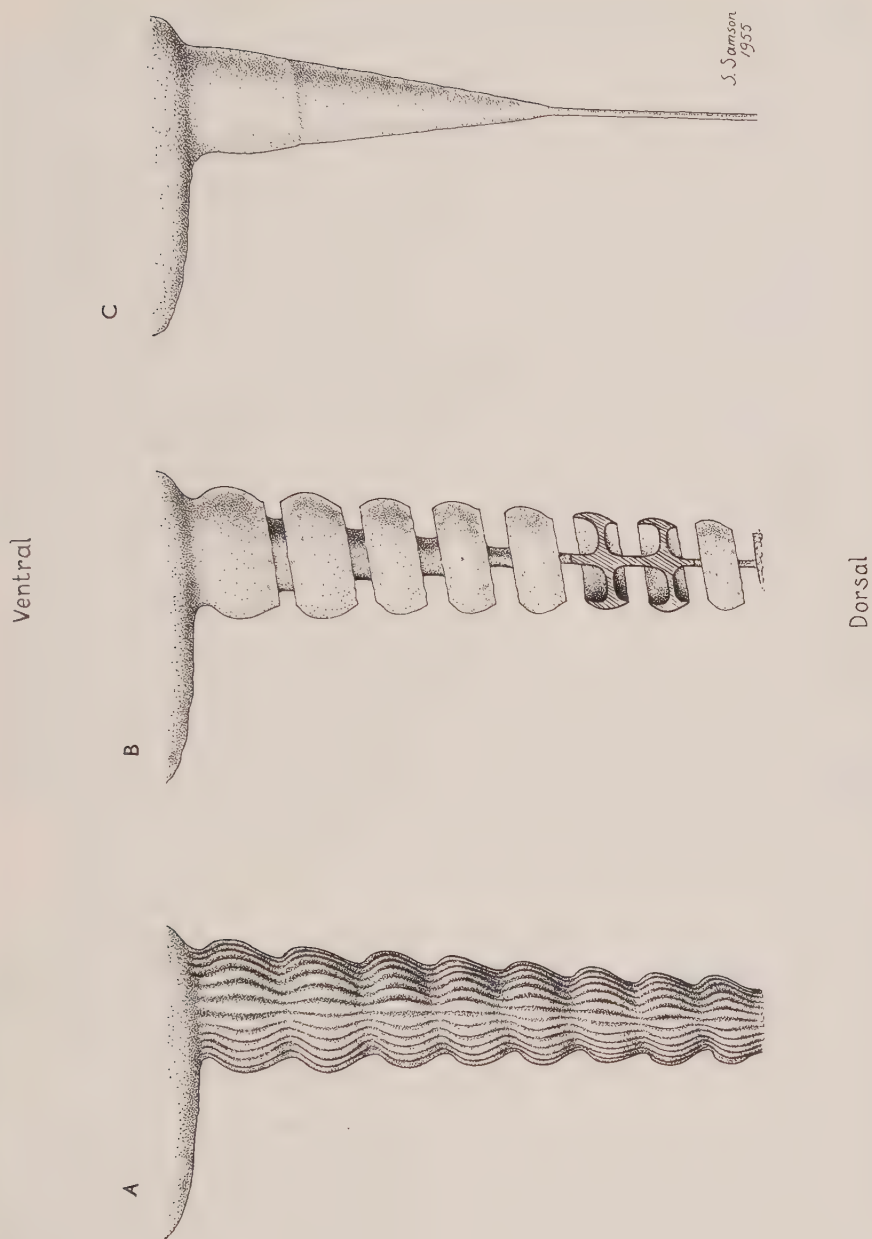


Fig. 4. Schematized reconstruction of the reduced siphonal cord in the three types of the secondary endosiphonal deposits. A, in the radial (Fig. 3 A); B, in the annular (Fig. 3 B); and C, in the conical (Fig. 3 C).

Precipitation of lime in the shell chambers of the fossil Nautiloids

In *Nautilus* and *Spirula* the shell chambers are filled by gas (Vrolik 1843; Willey 1902; Bruun 1943). As the majority of the writers have pointed out, this was certainly true also in fossil Nautiloids. When, after the death of the animal, the shells of these Nautiloids sank down on to a calcareous bottom sediment and were gradually embedded in it, the lime-saturated water above and about this sediment would percolate through the distal division of the siphonal funnel into the shell chambers. Thereafter the lime crystallized out on the inside of the shell wall and on the dorsal and ventral surface of the septum proper. Such a crystallization could under certain physico-chemical conditions produce lime precipitations of very peculiar form and structure, as e.g. the lamellar filling of the shell chambers in "*Lamelloceras*" spp. (Pl. 1; see Le Maitre 1950; Termier 1950 a, b), which I have investigated more closely.¹ The lime precipitations, formed in the way just described, have been misinterpreted by numerous writers (e.g. Barrande 1859, 1877a; Blake 1882; Hyatt 1900; Ruedemann 1906; Grabau 1922; Croneis 1926; Teichert 1933, 1934 a, b, 1935; Teichert & Glenister 1952, 1953; Schindewolf 1935, 1942; Miller, Dunbar & Corda 1933; Kobayashi 1936 a, b, 1937; Flower 1936, 1939 a, b, 1940, 1942, 1943a, 1946, 1950, 1955a; Ulrich, Foerste, Miller & Unklesbay 1944; Termier 1950 a, b) as secretions of the tissues of the animal. More exactly, these precipitations, which have been called "dépôt organique dans les loges aériennes" (Barrande), "intrakamerale Ablagerung" (Teichert, Schindewolf), "intracameral stereoplasmic deposit" (Kobayashi), "cameral deposit" (Flower) etc., should have been formed either by the epithelium on the dorsal surface of the body proper (Barrande) or by special tissues in the shell chambers which were completely separate from the tissues of the rest of the body and called by Flower "cameral mantle" (Teichert and Flower). Both these interpretations are, however, quite erroneous and have arisen owing to the imperfect knowledge of the above-mentioned writers of the anatomy of Cephalopods and of Molluscs in general. This is also true of Teichert's (1933) and Flower's (1939 b, 1946, textfig. 2) statements that living tissue would be found in the shell chambers of *Nautilus*.

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¹ These forms belong to a large collection of material of Paleozoic Nautiloids which I had opportunity of collecting in French North Africa through the generous support of "Centre de Recherches Sahariennes", "Service Géologique du Maroc" and "Direction générale des Relations culturelles du Ministère des Affaires étrangères", and I would like here to express my very sincere gratitude to the Directors of these Institutes. I wish also to record my best thanks to Prof. P. Pruvost and to Dr J.-P. Lehman who helped in every possible way.

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Plate I

Figs. 1, 2, 3, 4. Sagittal section through the septate division of the shell in four undetermined Nautiloids, probably of the same genus and species (= "*Lamelloceras*"). These figures illustrate the variation of the post-mortem lime precipitation in the shell chambers. N. Africa, N. Sahara, Marhouma (Saoura), Eifélien, Devonian. $\times 5/4$ of the natural size.

Tryckt den 5 december 1956

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Utgivet den 25 januari 1957.